4th International Workshop on
Scattering of Atoms and Molecules from Surfaces (SAMS-4)
Madrid, September 24th–27th, 2019

BOOK OF ABSTRACTS

Support has been provided by the US Army CCDC-Atlantic and the US Air Force EOARD
Welcome to Madrid, Spain, to the 4th International Workshop on Scattering of Atoms and Molecules from Surfaces, SAMS-4.

SAMS is the acronym for "Scattering of Atoms and Molecules from Surfaces". The previous three meetings of this series were held in Israel (2010), Germany (2013) and Norway (2016). The purpose of the fourth edition of this Workshop is to bring together scientists working in the general field of atom/molecule surface scattering. Participants include a balanced mix of the leading experimentalists and theorists in the field. The central aim of the workshop is to discuss most recent advances in both experiment and theory, stressing the unsolved and challenging problems which will be faced in the near future.

We look forward to a wonderful week of lively discussions and hope that you will enjoy your stay in Madrid.

Daniel Farías, Universidad Autónoma de Madrid
Salvador Miret-Artés, Instituto de Física Fundamental, CSIC
Program
FOURTH INTERNATIONAL WORKSHOP ON
SCATTERING OF ATOMS AND MOLECULES FROM SURFACES

FINAL PROGRAM

TUESDAY, SEPTEMBER 24

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WEDNESDAY, SEPTEMBER 25

**2D Materials**

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“Surface structure and dynamics of topological insulators from helium atom scattering”

11:30  A. Ruckhofer  
“The Atom-Surface Interaction Potential of Topological Insulators from Helium Scattering”

**Quantum Scattering**

11:50  E. Pollak  
“Theory and Manifestations of Quantum Reflection”

12:30  B.S. Zhao  
“Matter-Wave Diffraction from a Periodic Array of Half Planes”

12:50  LUNCH BREAK

**Metals**

14:30  M. Alducin  
“Ultrafast dynamics of CO adsorbates on metal surfaces under thermal and non-thermal conditions”

15:10  J. Subotnik  
“Different ways of understanding dynamics at surfaces”

15:50  H. Ueta  
“Rotational-state distribution and ortho-para conversion of molecularly chemisorbed H₂ on Pd(210)”

16:10  D. Farías  
“Diffraction of CH₄ from a Metal Surface”

16:30  A. Rivero  
“Ab initio Molecular Dynamics Study of alignment resolved O₂ scattering from highly oriented pyrolytic graphite”

16:50  Poster Session & Coffee break
THURSDAY, SEPTEMBER 26

Hydrogen & Oxygen Scattering

9:00  K. Fukutani
“Rotational dynamics of molecular hydrogen on a surface and in scattering”

9:40  S. Sibener
“Spatio-Temporal Visualization of Interfacial Chemical dynamics via Combined Gas-Surface Scattering and STM/AFM Imaging”

10:20  Coffee break

10:50  H. Chadwick
“Using Rotational Waves to Probe Gas-Surface Dynamics”

11:10  J.R. Manson
“Effect of static surface disorder on the diffraction of H$_2$ from Ru(0001)”

Graphene & Graphite

11:30  G. Benedek
“The electron-phonon coupling strength of 2D materials from He atom reflectivity”

12:10  A. Wodtke
“Imaging covalent bond formation by H-atom scattering from Graphene”

12:50  LUNCH BREAK

14:30  M. Hernández
“Interaction and dynamic between rare gases and 2d layered materials”

Dynamics & Diffusion

15:10  R. Marquardt
“The Diffusion Rates of Adsorbates from Time Dependent Quantum Dynamics”

15:50  A. Tamtoegl
“The Range of dynamics Studied with Atom-Surface Scattering: Verifying Rate Theory Across 14 Orders of Magnitude”

16:10  Coffee break
16:40  Q. Shi  
“Charge transfer and vibrational relaxation at surfaces: Theoretical studies based on non-perturbative reduced dynamics”

17:20  P. Fouquet  
“Surface friction processes revealed by a combination of neutron and helium atom scattering”

21:00  Conference dinner

FRIDAY, SEPTEMBER 27

Neutral Helium Microscopy

9:00  A.P. Jardine  
“Contrast in Scanning Helium Microscopy: New Opportunities and Challenges in Atom Surface Scattering”

9:40  S.D. Eder & M.G. Barr  
“Fundamentals of imaging in the scanning helium microscope: image formation, contrast mechanisms, and applications”

10:20  N. Avidor  
“Towards Motion Based Chemical Contrast in Helium Scattering”

10:40  Coffee break

Helium Atom Scattering

11:10  P. Roncin  
“Inelastic diffraction of fast atoms on surfaces, Lamb-Dicke regime, local Debye model and phonons modes”

11:30  C. Díaz  
“Prominent out-of-plane diffraction in Helium scattering from a methyl-terminated Si(111) surface”

11:50  A. Muzas  
“PSMS-CRP reactive potential energy functions: from frozen to mobile surface atoms”

12:10  Concluding Remarks
Sep. 24, TUESDAY
Overview of SAMS4: Scattering at Surfaces – Quo vadis?

W. Allison

*Cavendish Laboratory, University of Cambridge, Cambridge CB30HE*

SAMS meetings highlight work where scattering from surfaces is both a central topic of interest and the tool by which other surface processes can be investigated. An example of the former lies in understanding the quantum dynamics of molecular scattering, while the use of scattering to investigate surfaces is pre-eminently illustrated by helium atom scattering (HAS). In both areas mature techniques sit alongside newer innovations and, importantly, progress follows from a combination of experiment and closely developed theoretical understanding.

Some highlights in recent developments will be described alongside areas with potential for deeper understanding that will broaden the area of application for scattering data and possibly widen the scientific base in which we work. In the present overview I hope to set out a positive perspective on future developments, where a more holistic treatment of experimental data will be key.
Two pieces of system-bath dynamics for energy transfer at surfaces

Peter Saalfrank

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Energy transfer between an adsorbate (a “system”) and a solid surface (a “bath”) is a central issue in many reactive and non-reactive gas-surface encounters. In this talk, two selected examples of ultrafast “system-bath” dynamics at surfaces will be addressed.

In a first example, the vibrational relaxation of adsorbates by vibration-phonon coupling will be treated by various quantum mechanical approaches: Multi-dimensional system-bath Schrödinger equations, solved by Multi Configurational Time Dependent Hartree (MCTDH) or by the so-called Bixon-Jortner methods, will be compared to time-dependent perturbation theory and Lindblad open-system density matrix approaches [1,2,3].

In a second example, the coupling of nuclear adsorbate degrees of freedom to electron-hole pairs and “hot electrons” in a metal surface will be treated, for the example of CO on transition metals (Ru(0001) [4,5] and Cu(100) [6]). Specifically, inelastic scattering as well as femtosecond-laser induced processes will be considered. In these cases we use classical dynamics on ab initio potential energy surfaces, and the coupling to an environment is described within a Langevin formalism.

References
Save Helium Atom Scattering

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\textsuperscript{2}The Cavendish Laboratory, University of Cambridge, JJ Thomson Avenue, Cambridge, United Kingdom

In this presentation we will discuss the critical situation of helium scattering instrumentation including helium spin echo. The number of groups working with the techniques is reaching a point which is close to being "below critical mass". Because the techniques are not commercially available they are little known and little used in the scientific community at large and it is difficult to get access to experimental facilities for those who might be interested. This is a problem for the scientific community in general, and the material science community in particular, because helium scattering including helium spin-echo scattering can provide the answers to some scientific questions which cannot be answered with other techniques. We scientist working with helium scattering techniques have an obligation to the scientific community at large to ensure that these techniques will be available for future generation. The question is what to do? In this talk we will present a vision of setting up a small number of international centres for helium scattering that can facilitate external requirements. It may be natural that at least one of these centres is located at a big research facility, for example the new Neutron facility in Sweden. We note that user friendly data analysis software will also be called for.

Cambridge has received a grant to make the spin-echo facility available for external users in the next few years. The setup will be presented in more detail in the presentation. This can provide an important testing ground for the international centre idea. The presentation will open for a broad discussion.
Sep. 25, WEDNESDAY
Low-energy plasmons in Dirac materials

Antonio Politano

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The advent of graphene \(^{[1]}\) and other layered materials “beyond graphene” \(^{[2]}\) has opened the way for plasmonic applications covering a wide frequency range, ranging from terahertz (THz) up to the visible. In particular, the two-dimensional electron gas (2DEG) supported by Dirac-cone electrons enables gapless intraband plasmon in doped graphene \(^{[1]}\) and topological insulators \(^{[3]}\). Experiments by angle-resolved high-resolution electron energy loss spectroscopy have evidenced that the plasmonic mode of Dirac fermions (Dirac plasmon) in Bi\(_2\)Se\(_3\) topological insulator \(^{[4]}\) is strongly hybridized with the surface plasmons arising from three-dimensional electron gas associated to the bulk concentration of charge carriers.

The dispersion relation of Dirac plasmon is modified by the presence of a metal substrate (as in the case of graphene/metals \(^{[5]}\) or a metal gate \(^{[6]}\). In field-effect transistors (FETs) with an active channel of graphene \(^{[7]}\) or topological insulators \(^{[8]}\), it is possible to excite acoustic plasma waves (overdamped at room temperature), used for rectification of THz light. Plasma-wave THz photodetection has been demonstrated in antenna-coupled FETs with active channel of Bi\(_2\)Te\(_{2.2}\)Se\(_{0.8}\) \(^{[8]}\). These plasmonic THz photodetectors have been used for imaging experiments in the prospect of novel THz body scanners for homeland security \(^{[8]}\).

Dirac semimetals, in which bulk states form a Dirac cone, show different plasmonic modes. In type-II Dirac semimetal PtTe\(_2\), a gapped dispersing plasmonic excitation exists (~0.5 eV at Γ), with an anisotropic plasmon gap associated to the anisotropy of the tilted Dirac cone \(^{[9]}\).

Finally, surface plasmons of single crystals of NbAs and TaAs Weyl semimetals \(^{[10]}\), with frequency tunable by chemisorbed species \(^{[11]}\), will be presented.

References

\(^{[1]}\) A. Politano, G. Chiarello, Nanoscale 2014, 6, 10927-10940.
\(^{[2]}\) A. Agarwal, M. S. Vitiello, L. Viti, A. Cupolillo, A. Politano, Nanoscale 2018, 10, 8938-8946.
\(^{[3]}\) A. Politano, L. Viti, M. S. Vitiello, APL Materials 2017, 5, 035504.
\(^{[7]}\) A. Tomadin, M. Polini, Phys. Rev. B 2013, 88, 205426.
Chemically stable bilayers of crystalline and vitreous SiO$_2$ (2D silica) represent a new class of wide band gap 2D material [1]. From a mechanical point of view the flexibility of 2D layers is a big advantage for a range of applications, for example flexible electronics, where both conductive and insulating layers are needed. Furthermore it may be possible to tune properties such as band gap, thermal conductivity and resistivity through strain engineering. One important mechanical property is the so called bending rigidity, known also as flexural bending rigidity, flexural rigidity, bending modulus or bending stiffness.

Up till now graphene is the only 2D material where the bending rigidity has been measured directly experimentally, with big deviations between theory and experiments for the initial measurements [2,3]. In 2015 Al Taleb et al introduced helium atom scattering (HAS) as a new technique for measuring the bending rigidity of 2D materials. They extracted the bending rigidity from the low-energy phonon modes and obtained the first experimental value in agreement with theory [4]. The classical formula for bending rigidity of a plate is dependent on Young's modulus and Poisson's ratio and the thickness of the material. Several experiments exist on the measurements of elastic moduli [5] and it should thus be possible to obtain information about the bending rigidity from the elastic moduli measurements and vice versa. However, this implies knowing the thickness. For graphene the thickness value has proven to be ambiguous with different plausible values giving very different values for Young's modulus for carbon nanotubes. This is known as the Yakobson Paradox. See for example [6,7]

Here we use HAS to obtain the first measurement of the bending rigidity of a non-monoatomic 2D-material - 2D silica [8]. We compare our results with DFT calculations and obtain good agreement. We also compare our results with the classical formula for bending rigidity and discuss the implications for Yakobsons paradox.

References:
Surface structure and dynamics of topological insulators from helium atom scattering

A. Tamtögl¹, A. Ruckhofer¹ and W. E. Ernst¹

¹Institute of Experimental Physics, Graz University of Technology, 8010 Graz, Austria

Materials with peculiar electronic surface effects such as the novel group of topological insulators are particularly interesting for truly surface sensitive measurement methods such as helium atom scattering (HAS). Using HAS we were able to obtain accurate He-atom-surface interaction profiles by analysing ultrahigh resolution measurements of selective adsorption resonances and determine accurate electronic surface corrugation for Bi²Te３[1], Bi₂Se₃, and Sb₂Te₃. By adapting a recently developed quantum-theoretical derivation of the helium scattering probabilities to the case of degenerate semiconductors [2], we can extract the electron-phonon (e-ph) coupling parameter λ from the Debye-Waller attenuation of the elastic diffraction peaks [3].

Inelastic scattering experiments provide access to surface phonon dispersion curves that we compare with density functional perturbation theory calculations [4] which show excellent agreement. Besides the expected modes, the experimental data reveal two additional dispersion curves in the gap below Rayleigh waves which will be discussed in the talk.

Figure: Helium atom – surface interaction potential from the analysis of selective adsorption resonances.

Acknowledgments: This work has the support of the Austrian Research Fund FWF.

References
The Atom-Surface Interaction Potential of Topological Insulators from Helium Scattering

A. Ruckhofer\textsuperscript{1}, M. Pusterhofer\textsuperscript{1}, M. Bremholm\textsuperscript{2}, W. E. Ernst\textsuperscript{1} and A. Tamtögl\textsuperscript{1}

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\textsuperscript{2}Centre for Materials Crystallography, Department of Chemistry and iNANO, Aarhus University, Aarhus, Denmark

Helium atom scattering permits measurements of the atom-surface interaction potential to a very high accuracy via selective adsorption resonances (SARs) \cite{1,2}. Studying SARs provides access to the bound state energies which are supported by the atom-surface interaction potential. He atom scattering spectra of Bi\textsubscript{2}Se\textsubscript{3} and Sb\textsubscript{2}Te\textsubscript{3} show several SAR features which were analysed, starting with the free-atom approximation and a laterally averaged atom-surface interaction potential. Based on quantum mechanical calculations of the He-surface scattering intensities and resonance processes the three-dimensional atom-surface interaction potential is considered which is further refined in order to reproduce the experimental data\cite{1-4}.

The results are compared with previous measurements of Bi\textsubscript{2}Te\textsubscript{3}\cite{2} and allow for a comparative study of archetypal two-component topological insulator surfaces regarding their potential shape and the surface electronic corrugation. In addition, the angular broadening of SARs in experimental measurements is related to the lifetime of the corresponding bound states, providing an estimate of the time the He atom stays in the bound state before it leaves the surface.

The presented results may also be used to capture the fundamentals of adsorption systems where the binding is governed by long-range electronic correlations\cite{5}: Since the meV He beam is scattered in the low density region dominated by the tails of Fermi level surface states, studying SARs provides access to the interaction of TI surfaces within the weak adsorption regime.

![Figure 1: Illustration of a resonance process in which the He atom is trapped in a bound state of the interaction potential.](image)

References

Quantum reflection is complementary to quantum tunneling, yet the attention and understanding of quantum reflection is arguably less developed. A striking result is in the observations of scattering of He dimers from a surface at incident grazing angles [1]. Quantum reflection prevents the very weakly bound dimer from coming close to the surface and therefore it is not necessarily dissociated upon scattering. Yet the theory of quantum reflection from surfaces has been clarified only in recent years. It is well understood that the Casimir van-der-Waals long-range interaction potential has a region in which at very low energies creates a region in which the deBroglie wavelength of the incident particle changes rapidly. In this so-called “badlands” region the semiclassical approximation breaks down. This region occurs far from the surface and has been suggested to lie at the heart of the quantum reflection phenomenon. In this talk, we will show [2] that the badlands region is not important, but rather the essence of quantum reflection lies in the extremely long wavelength of the incident particle. Moreover, using close coupled scattering computations for surface scattering one finds that quantum reflection is influenced by the global potential energy surface and not only by a localized badlands region [3].

Even the basic theory of quantum reflection by a one-dimensional barrier potential leads to surprises. In a detailed study of quantum reflection [4] from a square barrier we show that there is a transition from incoherent to coherent quantum reflection as the incident energy is lowered. In the coherent regime we find that the “reflection time” is not given correctly when using the Wigner phase time delay expression.

Another manifestation of quantum tunneling is the long time dependence of wavepacket scattered from potential surfaces. In contrast to a free particle where the long time tail goes as $t^{-1}$ quantum reflection creates a long time tail that goes as $t^{-3}$. Finally, we show that quantum reflection can under certain circumstances lead to an inverse isotope effect, by which a hydrogen atom will diffuse slower than a D atom.

References
Matter-Wave Diffraction from a Periodic Array of Half Planes


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We report on reflection and diffraction of beams of He and D$_2$ from square-wave gratings of a 400-µm period and strip widths ranging from 10 to 200 µm at grazing-incidence conditions. In each case we observe fully resolved matter-wave diffraction patterns including the specular reflection and diffracted beams up to the second diffraction order. With decreasing strip width, the observed diffraction efficiencies exhibit a transformation from the known regime of quantum reflection from the grating strips [1, 2] to the regime of edge diffraction from a half-plane array [3]. The latter is described by a single-parameter model [4] developed previously to describe phenomena as diverse as quantum billiards, scattering of radio waves in urban areas, and reflection of matter waves from microstructures. Our data provide experimental confirmation of the widespread model. Moreover, our results demonstrate that neither classical reflection nor quantum reflection are essential for reflective diffraction of matter waves from a structured solid, but it can result exclusively from half-plane edge diffraction. We apply diffractive reflection also to fragile He$_2$ and He$_3$ clusters. Unlike He atoms, the clusters’s diffractive reflection probability is significantly increased as compared to quantum reflection.

Figure: (a) Schematic of the experimental setup. The grating normal is chosen as the z axis of our coordinate system. Incidence $\theta_{in}$ and detection angle $\theta$ are measured with respect to the grating surface in the $xz$ plane of incidence. (b) The reflection gratings are 50-mm-long microstructured arrays of 4-mm-long parallel strips made out of 1-µm-thick photoresist patterned on a commercial gold mirror. The center-to-center distance of the strips defines the period $d = 400$ µm identical for all gratings used. Four gratings with strip width $a = 10, 30, 100,$ or 200 µm have been used. For all four gratings, the gold surface between the strips is completely shadowed by the strips for all incidence angles used in this work. A sketch of the $a = 10$ µm grating is shown in (b). (c) Representative diffraction pattern for $a = 10$ µm and $\theta_{in} = 0.984$ mrad measured by rotating the detector around the y axis and integrating the signal for 8 s at each angular position.

Acknowledgments: This work has the support of the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT, and Future Planning (Nos. NRF-2015R1A2A2A01005458, NRF-2017R1A2B4008226, and NRF-2017H1A2A1042369).
References
Ultrafast dynamics of CO adsorbates on metal surfaces under thermal and non-thermal conditions

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$^2$Donostia International Physics Center, Donostia-San Sebastián, Spain

An important challenge in surface chemistry is to understand the details of surface reactions in both time and space domains. Experimentally, pump-probe time-resolved vibrational spectroscopy is among the few techniques that permit to monitor the initial steps and evolution of the adsorbate dynamics. In many cases, it is the CO internal stretching (IS) vibration the property that serves to probe the adsorbate dynamics. However, the experiments alone cannot provide a precise understanding of the mechanisms responsible for the frequency and linewidth changes that are observed under nonthermal conditions or, simply, the mechanisms behind the short IS lifetime also measured at equilibrium.

In this talk, I will introduce two different theoretical approaches we have followed to treat the complex nonadiabatic dynamics of CO on metals. The first one is based on many-body perturbation theory and we have applied it to investigate the lifetime of the IS mode of CO adsorbed on Cu(100) [1], as well as the transient frequency and linewidth changes that are measured when the adsorbed CO is exposed to the extreme nonthermal conditions initiated by femtosecond laser pulses [2]. Our calculations reveal in both cases that electrons, besides coupling directly to the vibrating CO, drive the otherwise forbidden coupling between modes with very different energies [1,2]. An alternative approach we have used to track the full dynamics of CO desorbing from Pd(111) consists in performing classical ab initio molecular dynamics with electronic friction (AIMDEF) [3,4,5]. Using this approach, we determine the reasons behind the experimental strong coverage dependence of the CO desorption yield. We also identify how the hot electrons and phonons cooperate in the desorption process [6].

Acknowledgments: Work developed with Spanish (FIS2016-76471-P), European (KK.01.1.1.01.0001), and German (TR1109/2-1) financial support.

References
Different ways of understanding dynamics at surfaces

Joseph Subotnik
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The theory of molecular scattering events at a metal surface is one of continuing fascination for chemists and physicists alike; there are just so many ways to exchange energy. One can observe electron transfer, excitation energy transfer with electron holes pairs, vibrational energy transfer, spin crossovers, and more. In this talk, I will highlight our recent attempts to understand these dynamics through different techniques valid in different regimes: electronic friction as valid in the adiabatic limit, surface hopping as valid in the nonadiabatic regime. I will highlight several open questions as to just how these methods can be used in practical calculations on surfaces, and I will also (time permitting) discuss the interesting question of how continuous wave illumination would affect the relevant scattering processes.
Rotational-state distribution and ortho-para conversion of molecularly chemisorbed H$_2$ on Pd(210)

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Imperfections of the surface, such as steps, are known to affect adsorption and reaction processes. While hydrogen typically chemisorbs dissociatively on a flat metal surface, it is known that hydrogen can molecularly chemisorb on stepped surfaces, such as Ni(510)$^1$ and Pd(210)$^2$, in which the potential is expected to be highly anisotropic, unlike the case of physisorption. The anisotropic potential would modify the rotational state of H$_2$ and promote ortho-para (o-p) conversion. For the system of H$_2$/Pd(210), the conversion time of ~1 sec was predicted based on the measurement on rotational-state-selective thermal desorption of H$_2$ ($J=0$ and $J=1$) $^3$. The predicted value is indeed much shorter than those in previously reported physisorption systems $^4$.

In this contribution, to further probe the rotational state and conversion of molecularly chemisorbed H$_2$, a combination of a pulsed molecular beam, photo-stimulated desorption and resonance-enhanced multiphoton ionization techniques is used. We will present the rotational state distribution of photo-desorbed H$_2$, and the o-p conversion process of molecularly chemisorbed H$_2$ will be discussed.

Acknowledgments: This work was supported by JSPS KAKENHI Grants JP17H01057 and JP18H05518, and Leading Initiative for Excellent Young Researchers, MEXT, Japan.

References

Diffraction of CH$_4$ from a Metal Surface

Amjad Al Taleb$^{1,2}$, Gloria Anemone$^1$, Linsen Zhou$^3$, Hua Guo$^3$, and Daniel Farías$^{1,4}$

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Diffraction with matter waves has been reported since the beginning of quantum mechanics. In free space, diffraction effects have been observed even with objects as large as C$_{60}$ molecules [1]. However, in scattering from a solid surface, pure elastic diffraction features have never been observed with molecules larger than D$_2$ [2]. Here we report the first observation of pure molecular diffraction for CH$_4$ scattered off of an Ir(111) surface [3]. These results prove that quantum coherence is preserved, despite the small separation between rotational levels and the interaction with surface phonons. Density functional theory calculations of the potential energy surface provide some clues to understand the larger corrugation sampled by CH$_4$ molecules in comparison to Ne atoms. Measurements of similar data for other polyatomic molecules will allow characterization of the multidimensional PES with unprecedented precision.

Figure: Angular distributions of CH$_4$ scattered from Ir(111) along the ΓM direction. The surface temperature is 110 K, and the beam energy 81 meV. Vertical lines indicate the expected position according to the Bragg condition [3].

References
Ab Initio Molecular Dynamics Study of alignment resolved O2 scattering from highly oriented pyrolytic graphite

A. Rivero Santamaria\textsuperscript{1}, M. Alducin\textsuperscript{1,2}, R. Díez Muiño\textsuperscript{1,2} and J. I. Juaristi\textsuperscript{1,2,3}

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In order to contribute to the theoretical understanding of recent observations\cite{1} on the alignment resolved O\textsubscript{2} scattering from highly oriented pyrolytic graphite (HOPG), we have performed \textit{ab initio} molecular dynamics (AIMD) calculations. The Vienna Ab Initio Simulation Package (VASP)\cite{2,3} was used to carry out spin polarized calculations using the generalized gradient approximation (GGA) by Perdew-Burke-Ernzerhof (PBE)\cite{4}.

The results obtained from simulations of a molecular oxygen beam with a selected orientation impinging on a thermalized graphite surface at 110 and 300 K, with an incident energy of 200 meV, show that the long range forces dominate the O\textsubscript{2}-HOPG interaction. The AIMD scattering “total probabilities” are almost independent of the initial orientation of the molecule and surface temperature, when only “in plane” scattered molecules are taking into account, the differences in the probabilities between molecules with parallel and normal orientation is enhanced. The energy loss in the collision depends of the initial orientation of the O\textsubscript{2} molecule, the internal energy of incident perpendicular molecules is six times greater than parallel ones, pointing out that for end-on collisions the transfer of energy to the internal modes is more important than the transfer of energy to the surface.

References
\begin{itemize}
\item \cite{1} Kurahashi, M.; Takahiro, K, Phys. Rev. B. \textbf{99}, 045439 (2019)
\item \cite{2} Kresse, G.; Furthmuller, Phys. Rev. B. \textbf{54}, 11169 (1996)
\end{itemize}
Sep. 26, THURSDAY
Rotational dynamics of molecular hydrogen on a surface and in scattering

K. Fukutani$^{1,2}$

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Molecular hydrogen has a large rotational energy because of its small moment of inertia, therefore often retains its rotational motion in the adsorption state on a surface, being called a quantum rotor. Since the rotational symmetry is lost in the adsorption, the molecule-surface interaction is anisotropic depending on the molecular orientation, which modifies the rotational-energy state and affects its scattering. We study nuclear-spin state conversion [1], rotational dynamics [1,2], and molecular-hydrogen formation on solid surfaces. To this end, we develop molecular hydrogen and spin-polarized hydrogen beams combined with REMPI. In the present work, we report the present status of the spin-polarized hydrogen beam, and the rotational dynamics of molecular hydrogen in the adsorption state on Pd(210) and scattering on SrTiO$_3$(001).

When the Pd(210) surface is exposed to H$_2$ at ~30 K, H$_2$ is molecularly chemisorbed on a H-covered surface, which is revealed by a TPD peak at ~70 K. Rotational-state-selective detection of this TPD shows that the $J$=0 H$_2$ is desorbed at 74 K while the $J$=1 H$_2$ is desorbed at a higher temperature of 78 K [3]. This indicates that the degeneracy of the $J$=1 state is lifted by a large potential anisotropy. Theoretical calculations show that the rotational wavefunctions in both $J$=0 and 1 states are substantially distorted, which can be described by a sum of spherical harmonics in several $J$'s. To further analyze the rotational wavefunction, photodesorption of these H$_2$ states is conducted, where H$_2$ is desorbed in high $J$ states. The rotational-state distribution is found to compare well with that of the adsorption state obtained by the theoretical calculation. Knowing the surface potential is anisotropic, we discuss the possibility to separate the rotational state of H$_2$ by surface scattering. Within a first approximation, the scattering amplitude of a molecule at a surface is proportional to the square of the matrix element of the potential for each rotational state, and the anisotropic potential is effective for the $J$≥1 states. The scattering of H$_2$ in $J$=0 and 1 is investigated on SrTiO$_3$(001), and it is found that the intensity in $J$=1 is increased by 4% while that in $J$=0 is decreased by 14%, which is in agreement with theoretical calculations [4].

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References
Spatio-Temporal Visualization of Interfacial Chemical Dynamics
via Combined Gas-Surface Scattering and STM/AFM Imaging

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The scattering of atomic and molecular beams from well-characterized surfaces is an incisive method for studying the dynamics of gas-surface interactions, providing precise information on energy and momentum transfer as well as complex reaction mechanisms. Scanning probe measurements provide a powerful complement to scattering data as SPM measurements give a direct route to the visualization/spectroscopic characterization of interfacial atomic and nano systems. Such combined molecular beam/SPM measurements allow for full elaboration of the detailed spatio-temporal correlations that govern interfacial chemical dynamics and reactivity.

We will report at SAMS-4 on several endeavors involving precision gas-surface interactions. Our newest scattering instrument is now fully operational, combining in one facility both reactive scattering and time-sequenced STM/AFM visualization of reacting surfaces [1-3]. Our recent work on the oxidation of Si(111)-(7x7) will be presented as an illustration for examining reactivity at multiple binding sites in space and time. Next, the erosion and ablation of graphite when exposed to energy selected beams of molecular oxygen will be presented focusing on reaction probabilities and morphological evolution of the reacting HOPG interface [2,3]. We have found that different oxidation conditions produce morphologically distinct etching features: Anisotropic channels, circular pits, and hexagonal pits, and that the reaction mechanism is beam energy dependent and non-Arrhenius with respect to surface temperature. On this surface, we have also conducted scattering experiments with O2 and N2 where we monitor velocity and angle distributions for energy transfer for basal plane HOPG; this done in conjunction with MD simulations from the Hase group [4]. This section will conclude with our most recent findings on the energy-dependent oxidation of GaAs(110). If time permits, some final comments will be made on the enrichment and separation of isotopes [5] based upon gas-surface interactions.

Selected Recent Publications


Using Rotational Waves to Probe Gas-Surface Dynamics

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The rotational orientation of a molecule can affect the outcome of a gas-surface collision, for example it has been shown through desorption measurements that D$_2$ is more likely to dissociate on a Cu(111) surface when it is rotating like a helicopter (with the plane of rotation parallel to the surface) than like a cartwheel (plane of rotation perpendicular to the surface) [1]. Scattering experiments which study the effect of rotational orientation directly have been limited to a small subset of molecules; either those which are paramagnetic or can be prepared through laser excitation. Here, a new technique will be presented [2], which allows the rotational orientation of ground state molecules to be prepared, manipulated and controlled using a series of homogeneous and inhomogeneous magnetic fields. The first results that have been obtained using this method for the scattering of H$_2$ from LiF will be shown. Preliminary analysis of the data suggests that scattering from a LiF crystal can in itself produce a rotationally oriented H$_2$ beam, as predicted theoretically over 20 years ago [3]. The experimental results presented here provide a further, more stringent test of current state of the art theory, and will help to develop and benchmark theoretical models.

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References
Effect of static surface disorder on the
diffraction of H$_2$ from Ru(0001)

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Diffractive scattering of atoms and small molecules from surfaces is known to be very sensitive to details in the interaction potential energy landscape. The diffraction of H$_2$ from metal surfaces has been shown to be an indicator of the reactivity of the metal towards H$_2$. [1] However, a recent comparison of calculated and experimentally measured diffraction probabilities, [2] in which the theory was done with a sophisticated potential energy surface that accurately described sticking and dissociative reaction of H$_2$ to Ru(0001), showed substantial discrepancies in intensity with theoretical and experimental probabilities differing by factors of 2 to 3. [3,4] It is shown here that the assumption of a small amount of static surface disorder removes most of the discrepancies between experiment and theory. [5] In its simplest form the result is expressed in terms of a characteristic function depending on only a single parameter, the static mean square displacement.

References
The electron-phonon coupling strength of 2D materials from He atom reflectivity

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Helium atom scattering (HAS) has been theoretically demonstrated to be a sensitive probe of the electron-phonon (e-ph) coupling strength at metal and metal-overlayer surfaces [1-4]. In particular it was shown that the temperature dependence of the Debye-Waller exponent as derived from the thermal attenuation of He atom reflectivity provides a direct measurement of the e-ph mass-enhancement parameter $\lambda$ at conducting surfaces [4,5].

After briefly reviewing the results obtained for several metal surfaces and thin films [5,6], an extension of the theory to 2D materials will be presented and applied to the analysis of recent HAS reflectivity data for layer semimetal chalcogenides [7], topological insulators [8,9] and graphene on various metallic substrates [10]. The superior sensitivity of HAS to phonon-induced surface charge density oscillations also permits to unveil the occurrence of multivalley charge density waves, and eventually determine the critical exponents, at semimetal surfaces such as Sb(111) [9] and Bi(114) [11].

References
Imaging covalent bond formation by H-atom scattering from Graphene

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Viewing the atomic scale motion and energy dissipation pathways involved in forming a covalent bond is a long standing challenge for chemistry. Here, we perform scattering experiments of H atoms from graphene and observe a bimodal translational energy loss distribution. Using accurate first-principles dynamics simulations we show that the quasi-elastic channel involves scattering through the physisorption well where collision sites are near the centers of the six-membered C-rings. The second channel results from transient C-H bond formation, where H-atoms lose 1-2 eV of energy within a 10 fs interaction time. This remarkably rapid form of intramolecular vibrational relaxation results from the C atom’s re-hybridization during bond formation and is responsible for an unexpectedly high sticking probability of H on graphene.

Figure: Re-hybridization in the formation of a C-H bond in collisions of an H atom at a graphene surface. $H_Z$ and $C_Z$ are the distances of the H and C atoms from the graphene plane. Three trajectories are shown for H atoms with 1.92 eV incidence energy.

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0001912).
The interaction of atoms and molecules with carbonaceous layered materials is a topic of interest both from a fundamental and a more applied points of view.

We have been recently working on the construction of reliable global potential energy surfaces for these systems and their application in different dynamical processes. There is an ample literature in which these processes are studied by Molecular Dynamics (MD) simulations and/or approximate quantum models. In this contribution we report three-dimensional wave packet calculations to study the interaction potential between graphene and graphite with rare gas atoms as well as the transport of $\{4,3\}^\text{He}$ atoms through a periodic rigid membrane of graphdiyne -a novel material composed of nanopores at regular distances- and also through a holey graphene model (with a force-field based on simple Lennard-Jones interactions). A good agreement is found between approximate quantum models and present calculation for the case of graphdiyne, with a much more pronounced differences for the transmission through nanoporous graphene.

References:
The Diffusion Rates of Adsorbates from Time Dependent Quantum Dynamics

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A time independent quantum dynamical formulation of the dynamical structure factor (DSF) related to particle scattering at mobile adsorbates exists in the case when the adsobates’ excited states have finite lifetimes due to relaxation phenomena [1,2].

In ref.[1], the formula is evaluated quantum mechanically using wavefunctions, energies and lifetimes of vibrational states obtained for H/Pd(111) from first principle calculations. The results are capable of capturing qualitative features of diffusion rates measured by 3He Spin echo experiments [3].

In the present talk, I present and discuss a time dependent formulation which will directly link the motion of wave packets and the intermediate scattering function (ISF) [2,4].

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References
The Range of Dynamics Studied with Atom-Surface Scattering:
Verifying Rate Theory Across 14 Orders of Magnitude

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Predicting the rates of dynamical processes based on accurate computational calculations is crucial for an understanding of the underlying principles of chemical reactions. However, the rate description of dynamics often suffers from the lack of experimental data which covers the entire range of relevant timescales and temperatures. Based on a study of the surface diffusion of cobalt phthalocyanine (CoPc, $C_{32}H_{16}CoN_8$) on a Ag(001) substrate, we demonstrate that microscopy provides information in the low temperature regime[1], while only inelastic atom-surface scattering[2] can follow the diffusive process at high temperatures. By combining both experimental techniques we are able to follow the rate of translational motion over 14 orders of magnitude ($10^{-4}$ s$^{-1}$ to $10^{10}$ s$^{-1}$).

Notably, scattering data taken at high temperatures makes a meaningful comparison of the pre-exponential factor in the rate description with theoretical predictions possible. The importance of the prefactor is often underestimated due to the above described lack of accurate experimental references, since it mathematically refers to extrapolating the rate to infinitely high temperatures. In addition to the fundamental interest, the approach promises to provide a route by which the factors affecting structural assembly in organic thin films can be explored and understood at a molecular level[3].

Another scattering technique, neutron scattering, can also be used to obtain information about surface diffusion in specific cases. It allows again for studies in the fast timescale / high temperature regime. Recent measurements promise to provide insight into subtle effects during surface diffusion, such as the role of surface symmetry or the contact angle between the adsorbate and the substrate.

Figure 1: A cobalt phthalocyanine molecule, adsorbed on a Ag(001) substrate.

References
Charge transfer and vibrational relaxation at surfaces: Theoretical studies based on non-perturbative reduced dynamics

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Charge transfer reactions at surfaces and interfaces play an important role in many chemical processes. A non-perturbative method is proposed to simulate heterogeneous electron transfer dynamics in systems described by the Newns-Anderson type of models. The coupling between the molecule and the continuum electronic states at the metal or semiconductor surface is represented using a set of effective modes, by employing an exponential expansion of the bath correlation functions. Numerical examples are then presented for applications to model systems including: (1) photo-induced charge transfer at the molecule-semiconductor interfaces; (2) heterogeneous electron transfer at the molecule-metal interface; (3) vibrational relaxation on a metal surface.

References:
Surface friction processes revealed by a combination neutron and helium atom scattering

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The study of friction and diffusion processes on molecular length scales requires experimental methods that deliver sub-nanometer spatial resolution at nanosecond time resolution. After decades of research in friction the range of available spectrometers with these characteristics is still very limited. Two non-destructive techniques cater particularly well for studying surfaces diffusion: neutron and helium atom scattering. In addition, a wide choice of high-surface-density materials is available now that can be studied by both methods in a complementary way. These materials have the added benefit, that they are widely applied in energy storage, sensing and filtering. Hence, research on these materials has strong potential technical impact.

Our research programme started with rather small and structurally simple molecules on carbon substrates, such as the aromatic molecule benzene adsorbed on the basal plane surface of graphite(0001) [1,2]. For benzene-graphite(0001) at sub-monolayer coverage, we now established the inter-molecular energy dissipation and the surface friction [2,3] using a range of neutron time-of-flight spectrometers, neutron spin-echo as well as neutron diffraction. The results led us to a surprisingly simple model: the inter-molecular friction can be quantitatively explained by a model of colliding cogwheels (or rough hard disks) that we had developed on the basis of the long established rough hard sphere model for molecular gases (the equivalent 3D model). The model explains the collisional friction or viscosity based on universal constants only and makes almost no assumptions about the system.

References:
Sep. 27, FRIDAY
Contrast in Scanning Helium Microscopy: New Opportunities and Challenges in Atom Surface Scattering

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Scanning helium atom microscopy (SHeM) \cite{1} is a new imaging technique that has been developed in recent years by combining the benefits of atom-surface scattering with spatial resolution. The method involves forming a helium microprobe by collimation or diffractive focusing of the beam, followed by raster-scanning over a sample and counting the atoms scattered into a particular solid angle. The low energies of thermal helium atoms make the method particularly valuable for studying insulating surfaces such as biological materials or polymers, or sensitive surfaces such organic electronic or light harvesting molecules.

To understand contrast formation in helium microscopy, we are able to draw on the extensive research activity in helium-surface scattering that has been developed over a number of years \cite{2,3}. In general, we expect (i) diffuse or topological contrast due to diffuse scattering from the local structure; (ii) Debye-Waller or chemical contrast due to inelastic interactions with the surface; and (iii) reflective, diffractive and interference contrast due to elastic scattering. However, in many microscopy applications the surface condition is less well defined than ultra-high vacuum surface-studies, providing both challenges and opportunities.

An overview of the helium atom microscopy technique and its context will first be given, followed by the implementation we have developed in Cambridge, U.K., in collaboration with the University of Newcastle, Australia. Recent applications of SHeM will be reviewed, along with the practical benefits compared to other potential approaches.

We will then discuss the forms of contrast seen to date, the evidence for which has been gathered using a combination of imaging, in-situ experimental profiling, and numerical ray-tracing simulations \cite{4}. We will show that most surfaces scatter with a cosine-like distribution that is centred on the surface normal (analogous to Lambert’s cosine law in optics), although the distribution is locally modified. The extent of the deviation from cosine will be discussed. In addition, we will show that contrast can be strongly enhanced on particular surfaces using diffractive scattering, which indicates the prospects for enhancement using other surface-scattering phenomena. Surface structure also leads to shadowing and diffuse illumination, which both mask and enhance detail in subsequent images.

Finally, the second generation of helium microscope being developed in Cambridge will be discussed, which aims to simplify and broaden the applicability of the technique, extend the range of experiments which can be performed and to establish the subsequent user-base and long-term viability of the technique.
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References
Fundamentals of imaging in the scanning helium microscope: image formation, contrast mechanisms, and applications

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In the last decade, multiple experimental systems capable of generating spatially resolved scans of a sample surface via neutral helium atoms have come online, representing a significant advance to the pursuit of routine atom imaging [1-3]. Neutral helium microscopy is of particular interest for working with delicate materials that may suffer degradation and even damage under other microscopies utilising charged particles or light [4]. As with all other microscopies, the specifics of image formation within the instrument are crucial to gaining a full understanding of the produced micrographs. Questions around the sources of the observed contrast further complicate the issue. From the many decades of work on Helium Atom Scattering (HAS), it is been theorised that contrast in neutral helium microscopy will arise not only due to the surface topography, but also its composition and arrangement [5]. However, the development of a complete understanding of the different contrast channels and their respective contributions to a collected micrograph is still an active topic of investigation and one of great importance to the field. With the nature of image formation and the contrast mechanisms being intricately related, teasing them apart is key to pushing our understanding of the technique further.

Here we present work detailing contrast and image formation as explored in the Scanning Helium Microscope (SHeM). In particular, the discussion will cover the basics of the projection distortion that arises due to the instrument scattering geometry; supra- and sub-resolution contrast formation; the effects of multiple scattering on the produced image; and the exciting possibility of chemical contrast. Extension of the aforementioned concepts (chemical contrast excluded) allowed for an iterative ray tracing Monte Carlo model replicating diffuse scattering from a sample surface to be constructed.

With the ‘soft’ nature of the He probe, the SHeM presents the possibility for completely damage free imaging, making it a uniquely suited instrument for a variety of delicate sample systems. We will show how the SHeM can be used for surface quality investigations in the nanoscale regime; how it is possible to record images of OPV (organic photovoltaic) materials without any surface coatings or damage to the polymer layers; as well as the application of SHeM to the field of taxonomy, utilising a stereophotogrammetry approach to draw qualitative taxonomical information from biological samples.

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Motion Based Chemical Contrast in Helium Scattering
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Helium Atom Scattering (HAS) offers some unique properties for studying surfaces, but is considered to lack general mechanisms for chemical contrast. In search for such a mechanism, I am exploring the idea of Motion Based Chemical Contrast (MBCC): the dynamics parameters of individual species may serve as surface-specific signature with which one can chemically analyse a multi-species system. Recent results from a computational study on bi-species system suggest that at dilute adsorbate densities, the inter-cell (“slow”) diffusion can be sufficient for resolving the partial density of adsorbate from the total line-shape, and at higher surface densities, the intra-cell motion, which is less sensitive to inter-adsorbate interactions, can reveal the partial density.

In the talk, I will introduce Helium Spin Echo spectroscopy [1,2] which enables us to measure the ultra-fast molecular dynamics at the surface. I will then present a few case-studies of single-species measurements, to demonstrate how and why the surface dynamics can be a useful fingerprint of the adsorbates. Lastly, I will discuss in more details results from my recent computational study which suggest that MBCC may indeed be feasible.

Inelastic diffraction of fast atoms on surfaces,
Lamb-Dicke regime, local Debye model and phonons modes
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Instead of using thermal energies at quasi-normal incidence to probe crystal surfaces, grazing incidence fast atom diffraction (GIFAD or FAD) uses keV atoms at grazing incidence (θ ~1 deg.). The full diffraction pattern can be recorded at once with very high efficiency on a position sensitive detector opening specific application such as the online monitoring of epitaxial growth in molecular beam epitaxy chamber [1,2].

The decoupling of the fast movement parallel to the surface from the slow motion perpendicular allows the elastic diffraction pattern to be interpreted as arising from an effective particle with wave-vector \( k_\perp = k \sin \theta \) [3]. The effective energy \( E_\perp = E \sin^2 \theta \) can easily be adjusted between few meV where van der Waals contribution is important (e.g. bound state resonances [4]) up to few eV where \( \lambda_\perp \) is only few pm allowing high topological resolution [5]. Such a small wavelength is less than the room temperature vibration amplitude of the surface atoms so that no coherent diffraction should be observed. However, in GIFAD, several surface atoms exchange momentum with the projectile so that the associated Debye-Waller factor is greatly reduced [6].

In the inelastic regime, the fast movement parallel to the surface has to be taken into account. Using a description where each surface atom is a quantum Debye harmonic oscillator, the elastic scattering appears as a specific Lamb-Dicke regime where the momentum exchanged with each surface atom is not enough to trigger a vibrational excitation [7]. The inelastic scattering profiles could be derived without need of the specific surface phonon modes. In principle, these modes could be included by projecting the momentum transferred to all surface atoms onto the phonon displacement field [8] but the lack of resolution in the energy loss does not allow identification of the modes which could well be associated with multiple excitations [9].

We will review the progress of grazing incidence fast atom diffraction at surfaces (GIFAD) over the past years and address ongoing questions and perspectives.

References
Prominent out-of-plane diffraction in Helium scattering from a methyl-terminated Si(111) surface

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Organic-terminated semiconductor surfaces have been widely studied during the last decades, because due to their electrochemical and oxidative stability are very suited for a number of useful applications$^1$. Among these surfaces, CH$_3$-Si(111) has received special attention as it provides good performance for properties such as suppression of surface reconstruction and resistance to oxidation$^2$. Taking advantage of the detailed knowledge available on this system$^3,4$, we have used it as a benchmark to study helium atom scattering (HAS) from a highly corrugated system.

To perform our analysis, we have computed two continuous potential energy surfaces (PESs) based on density functional theory, with and without taking into account van der Waals forces. And subsequently, we have carried out quantum dynamics of these PESs to obtain the simulated diffraction spectra. As it can be seen from the figure, we have found that most diffraction takes place out-of-plane, and this result does not depend on the specific features of the simulations, such as the inclusion or not of the van der Waals forces. We hope that this striking theoretical finding will encourage new experimental developments to confirm this previously unreported effect.

![Figure: Diffraction probability for CH$_3$-Si(111) as a function of the diffraction orders.](image)

Acknowledgments: This work has been partially supported by the MINECO project FIS2016-77889-R and the ARC Discovery project DP160100059.

References
The Periodic Surface Modified Shepard interpolation method (PSMSIM), in conjunction with the Grow algorithm, is a powerful technique to obtain accurate continuous representations of gas-surface potential energy surfaces (PESs). It has been used in the later years to study with success the dynamics of mono and diatomic molecules interaction with metallic and decorated non-metallic surfaces, e.g. H2 scattered from Cu(111), Pt(111) and CH3-Si(111) or He scattered from CH3-Si(111). However, in its previous implementation, the PSMSIM could not deal with the complications of explicit surface atoms mobility, something that has been shown to be important for the introduction of temperature effects in gas-surface systems.

In this contribution, we show two extensions of the PSMSIM. The first one allows to include explicit surface atoms displacements in the interpolation while strictly maintaining all the periodic properties of the PES. The second one is an improvement of the convergence of the method by the smoothing of the actual interpolated potential in the same fashion as in the Corrugation Reducing Procedure (CRP). Explicitly, we show how a previously frozen surface interpolated PES can be used as the starting point to construct its mobile surface version. On top of it, we show how the CRP improves the convergence of calculated reaction probabilities and other important observables, using as a source a model potential.

**Figure 1:** Calculated probabilities of different reaction paths as a function of the number of points included in the interpolated potential with CRP (right) and without it (left). Dashed lines mark the calculated probabilities of the model potential.
POSTERS
A user facility for following atoms on surfaces

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Since the observation of atoms, it has been a desire to see their dynamics; however, on the atomic scale processes occur quickly, often over the time scale of a billionth of a second. There are few tools that can access the relevant time and length scales and thus the details of how atoms and molecules move could only be guessed, leading to a lack of benchmarking for modeling and theoretical understanding of processes. Remarkably, it is now possible to follow the motion of atoms and molecules over these length and time ranges using the helium spin-echo (HeSE) technique [1,2]. The apparatus is unique and the equipment and infrastructure to support its use has been developed over many years in Cambridge where it’s been applied to a variety of systems [3,4] and applications.

We are now transitioning the equipment from a local resource into a productive, flexible user facility. We present in the current work, examples of recent results demonstrating the capabilities of the operation, an overview of the mechanisms for access to and services to support the experiments enabled by the facility. The primary element of the capability is the HeSE instrument, supported by a further atom diffractometer for preparatory experiments, user support including training and software is provided for. The equipment forms a globally unique tool which is wide applicability and interest and user applications are actively encouraged.

Acknowledgments: The apparatus has been developed and built with the help and support of many friends and collaborators too numerous to name.

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[2] Science 304, 1790 (2004);
Diffuse contrast observed in helium atom microscopy

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Atomic or molecular beams as used for spectroscopy, have been demonstrated to have application in real space measurement of delicate samples [1], for example in the taxonomy of animal and plant samples [2]. Since the first images published by Holst et. al. in 2008[3], and in reflection mode images that followed in 2011[4], contrast reflecting the topography of the sample surface has been observed, the latest results of which we present here. However, the detailed variation in contrast across an image is shown to be highly dependent on the angular intensity profile on scattering. We present simulated and experimental micrographs from technological samples, and comment on the comparison with scattering theory which is typically referenced to traditional surface science samples. Our results demonstrate a highly diffuse scattering that is consistent with a cosine distribution with intensity centred normal to the surface.

Scattering from deep topographic features is considered using simulations [5] and compared with data collected from a carefully manufactured sample, (shown in figure), which demonstrates the significance of multiple scattering in image formation. Proper understanding of such contrast features is important in the interpretation of data measured with atom beams, for example in porous structures, which we also present.

![Figure: Helium image of different aspect ratio trenches, depth increasing left to right.](image)

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**References**

Theoretical description of H atom scattering from tungsten

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Recently, detailed theoretical and experimental investigations of energy transfer between hydrogen atoms and late-fcc transition metal (111) surfaces have been made and on the basis of these experiments, it was possible to clarify the role of electron-hole pair excitation during the scattering process. [1-4] However, those investigative efforts excluded metals that crystallise in other crystal structures, because the theoretical methods in those investigations are based on Effective Medium Theory (EMT) [5], a theory that has been formulated to describe fcc metals and their alloys. Here, I present my first efforts to extend the EMT formalism to describe H atoms interacting with bcc metal surfaces. The obtained energy formula is fitted to DFT energies for H atoms at W(111). The obtained global EMT-PES is used to perform molecular dynamics simulations to investigate of how the scattering dynamics between H atom and metal surface is affected by the crystal structure and surface facet and surface temperature.

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References
The Mystery of CH₄ – Graphene Interaction

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We have recently reported the first observation of quantum-coherent scattering and even pure molecular diffraction of CH₄ from Ni(111) and Ir(111) surfaces [1,2]. A quite surprising result was the observation of coherent scattering at perpendicular incident energies as low as 20 meV, a value well below the depth of the physisorption well (~130 meV). Our analysis shows that the main hurdle to observe diffraction comes from the combination of a very wide physisorption well with a large mass of surface atoms.

Here we show CH₄ scattering measurements from a graphene (Gr) layer grown on Ni(111), whose effective atomic mass is enhanced by the strong Gr-Ni interaction. The measured angular diffraction spectra are broad, as expected for classical objects, with no quantum features. This is a quite surprising result, especially considering that clear diffraction features are observed with Ne beams, with nearly the same mass of methane. DFT calculations show that the physisorption well of CH₄ on Gr/Ni(111) is even slightly shallower than on Ni(111). This is consistent with the observation of clear diffraction peaks in the scattering of H₂ and D₂ from Gr/Ni(111) and Gr/Ru(0001). This means that the Debye-Waller model is not enough to explain the weird scattering process of CH₄ from graphene, and that an additional effect must be involved.

References
Experimental determination of electron-phonon coupling constant and thermal expansion of natural MoS$_2$

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We have studied the (0001) surface of 2H-MoS$_2$ by means of helium atom scattering (HAS). The electron-phonon coupling constant of this system has been determined by measuring the thermal attenuation of the specular peak at surface temperatures between 100 and 500 K. HAS diffraction also reveals a 3% planar dilation of the surface layer, while step interference indicates a slight contraction of the surface layer thickness. By employing a recently developed quantum-theoretical approach applied to the case of layered degenerate semiconductors, we find $\lambda = 0.40$ [1]. The present HAS data provide evidence for a surface enhancement of electron-phonon interaction and a possible relevant role of surface relaxation.

The pronounced out-of-plane features observed confirm the high corrugation of the 2D surface unit cell. The observation of diffraction features along the two main high symmetry directions allows determining the in-plane surface lattice constant with high accuracy. The measured lattice constant is $a = (3.25 \pm 0.05)$ Å. Within experimental error, the MoS$_2$ lattice parameter was found to remain constant in the temperature range between 90 and 522 K, in good agreement with previously reported DFT based calculations [2].

References

Modelling thermal reactions on surfaces: Transition state theory with dynamical corrections.

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Transition state theory (TST) is undoubtedly the most successful and widely employed theoretical approach for studying reaction rates involving species that undergo reaction at or near thermal-equilibrium conditions, for which direct dynamical calculations are often unfeasible. However, it is well known that TST gives an upper bound for the rate coefficients since it counts any crossing of the transition state as a reaction. This approximation can be corrected by calculating short time trajectories started at the transition state to obtain the so called dynamical correction factors \cite{1}. Among other reasons, recrossing can originate from the unseparability of the reaction coordinate from all other degrees of freedom. Another dynamical origin might be the excitation of electron-hole pairs, that are well known to happen when molecules interact with metal surfaces.

We study two different gas-surface reactions to evaluate the role of dynamical effects on the reaction rate constants, and therefore, to evaluate the validity of conventional TST. On one hand, a model system for which appears likely to have a strong electronic coupling: H\textsubscript{2} recombinative desorption on the Cu(111) surfaces. An accurate description of the electronic excitations is accomplished by \textit{ab-initio} molecular dynamics with electronic friction (AIMDEF) method \cite{2}. This method is based on the local density electronic friction approximation (LDFA) \cite{3}, which was shown to successfully model translational energy loss measurements for H colliding with Au(111) \cite{4}. Calculations show that non-adiabaticity do not affect the dynamical factors. However, increasing the temperature decreases the reaction constant up to a factor of 2. On the other hand, we study H adsorption on graphene standing on Pt(111), for which harmonic TST predicts high adsorption probabilities compared to the experimental measurements. Classical dynamics simulations are run on the EMFT-REBO PES that has been validated by comparison of experimental measurements and direct dynamical simulations \cite{5}. Preliminary results predict high corrections to the TST rate constant for this system.

References

\cite{1} C.H. Bennett, Alg. Chem. Comp. 46, 63–97 (1977).
Towards a specific reaction parameter density functional for H$_2$ + Ni(111): Comparison of theory with molecular beam sticking experiments

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Accurate energy barriers for rate controlling elementary surface reactions are key to understanding, controlling and predicting the rate of overall heterogeneously catalyzed processes. The specific reaction parameter approach to density functional theory (SRP-DFT) in principle allows chemically accurate barrier heights to be obtained for molecules dissociating on metal surfaces, and such accurate barriers are now available for four H$_2$-metal and three CH$_4$-metal systems. Also, there is some evidence that SRP density functionals (SRP-DFs) may be transferable among systems in which the same molecule interacts with a low index face of metals belonging to the same group. To extend the SRP database, here we take a first step to obtain an SRP-DF for H$_2$ + Ni(111), by comparing sticking probabilities ($S_0$) computed with the quasi-classical trajectory method with $S_0$ measured in several molecular beam experiments, using potential energy surfaces computed with several density functionals. We find that the SRP-DF for H$_2$ + Pt(111) is not transferable to H$_2$ + Ni(111). However, the PBE-vdW2 functional describes the molecular beam experiments on H$_2$ + Ni(111) we deem to be most accurate with chemical accuracy, and may therefore be considered a candidate SRP-DF for this system, of which the quality still needs to be confirmed through comparison with an experiment it was not fitted to. However, the different molecular beam sticking measurements we considered showed discrepancies with one another and with the theory for incidence energies > 0.2 eV, and it would be good if better defined and more accurate experiments would be done for these energies to resolve these differences.

Acknowledgement
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Reactive scattering of O$_2$ on Cu(111) and Cu$_{ML}$Ru(0001)

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The reactive scattering of O$_2$ on transition metal play a fundamental role in chemical reaction due to its key role in industrial catalysis, oxidation processes or the development of fuel cells$^{1,2,3}$. We have studied the reactive scattering of O$_2$ on clean Cu(111) and Cu$_{ML}$/Ru(0001) by means of classical and quasi-classical (including the zero-point energy of O$_2$) molecular dynamics. To perform our dynamics studies, following Refs.$^{4,5}$, we have worked within the Born-Oppenheimer approximation The electronic term was computed using density functional theory (DFT) within the generalized gradient approximation (GGA) using the RPBE functional obtaining 6D-PESs. The kinetic term of the nuclei was treated classically, considering the energy-exchange surface-molecule and surface-bulk by using generalized Langevin oscillators. In order to obtain observables that could be directly compared with experimental results, we have computed the averaged termal sticking probability of O$_2$ on Cu(111), Cu$_{1ML}$/Ru(0001) and Cu$_{2ML}$/Ru(0001). In the case of Cu$_{ML}$/Ru(0001), both surfaces give very good results in comparison with experiment$^6$, especially upon the inclusion of the zero-point energy, confirming the accuracy of our model (see Fig.). Whereas for O$_2$-Cu(111) a direct comparison is more questionable.

Figure: Averaged initial sticking probability of O$_2$ on Cu(111) and Cu$_{nML}$/Ru(0001) at $T_s=350$ K.

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References
Beyond the Static Corrugation Model
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Einsteinweg 55, 2333 CC Leiden, The Netherlands

Hydrogen dissociation on a Cu(111) surface is an important benchmark system with a wide range of available data. By applying the Specific Reaction Parameter (SRP) methodology to DFT, we are able to predict the H\textsubscript{2} dissociation probabilities with reasonable accuracy (< 43 meV error).[1] Previous work has shown this agreement with experiment can be further improved by including a model for surface temperature effects. One of these models is the Static Corrugation Model (SCM), where the static surface atoms are displaced from their ideal lattice positions based on the modeled surface temperature. [2, 3] To describe the effects of these displacements, a three-body (H-H-Cu) coupling potential is introduced. The expression of this coupling potential was fitted to a collection of SRP-DFT results.

In this work we aim to go beyond the Static Corrugation Model by also treating the Surface atoms dynamically. We wish to achieve this through the application of an Embedded Atom Method (EAM) potential, an approach which has been shown to successfully reproduce a wide range of surface and bulk properties.[4]

Initial work has been performed on attaining an analytical expression for each of the EAM potential components originally fitted by Sheng et al.[4], which has been designed and shown to correctly reproduce lattice constants, phonon dispersion curves as well as many other properties for bulk copper.

A fit of these potential components was needed, as neither our potential energy surface expression nor the SCM are available within the dynamics code often used for EAM potentials. This also provided the perfect opportunity to use analytical expressions, which should reduce the computational requirements of the components. Finally, it would also allow for later work to attempt the construction of a new EAM potential using DFT data obtained with the SRP functional. The quality of the fit is judged based on the reproducibility of several bulk properties, as shown in the figures.

Using snapshots of the surface configurations generated with our EAM potential, we are also able to verify the quality of the configurations generated in the earlier SCM works.

References
**Figure 1:** Lattice constant of bulk Cu at different simulated temperatures. Shown are the results computed within LAMMPS using the original potential as provided by Sheng et al. (Black), a linear fit to the results published by Sheng et al. (Green), results obtained using our fortran code with the analytical fit to the EAM potential (Red), and results obtained using LAMMPS with the analytical fit to the EAM potential (Blue).

**Figure 2:** Cohesive energy per atom in an ideal lattice for different lattice constants. Shown are the results computed within LAMMPS using the original potential as provided by Sheng et al. (Black), results obtained using our fortran code with the analytical fit to the EAM potential (Red), and results obtained using LAMMPS with the analytical fit to the EAM potential (Blue).
# TIME TABLE

<table>
<thead>
<tr>
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<th>Sep, 26 THURSDAY</th>
<th>Sep 27, FRIDAY</th>
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