Wednesday Afternoon, November 6, 2002

Surface Science
Room: C-108 - Session SS1-WeA

Gas-Surface Dynamics
Moderator: J.J. Boland, University of North Carolina at Chapel Hill

2:00pm **SS1-WeA1 Interaction of Gas-phase H Atom with Metal Surfaces : Cu(111) vs Pt(111)**, ***S.J. Lee***, *J. Kim, J. Lee*, Seoul National University, Korea

The gas-phase H atoms of thermal energy impinging upon metal surface undergo reflection, adsorption, or abstraction of a surface H atom. The probability of each process strongly depends on the inelastic nature of the H-metal surface collision. Inelastic energy loss via phonon excitation is expected to be quite inefficient because of the light mass of H atom. Recently, Nienhaus et al. (1) have experimentally shown that H atom collision with metal surfaces induces e-h pair excitation. Then, one can raise the following interesting questions; 1) Is e-h pair excitation a dominant inelastic loss mechanism in H-metal surface collision? 2) If it were the case, what would be the effect of electronic band structure of metal in its interaction with H atom? In order to gain further insight into these questions, we have measured and compared the initial sticking probability(S_0), saturation coverage(θ_s), and abstraction cross section(s_{abs}) for hydrogen atom at Cu(111) and Pt(111) at 90K. We measure S_0=0.72, θ_s=0.95ML, and s_{abs}=1.30Å for Pt(111) and S_0=0.45, θ_s=0.32ML, and s_{abs}=5.5Å for Cu(111). We interpret the results in terms of e-h pair excitation whose efficiency is determined by the electron density of state at the Fermi level.

2:20pm **SS1-WeA2 Anomalous Reactive Scattering: Hyperthermal Energy Collisions of State-selected Bromine Ions on Pt(111)**, *P.L. Maazouz*, ***M. Maazouz***, *D.C. Jacobs*, University of Notre Dame

The dynamics of scattering state-selected atomic and molecular bromine ions on Pt(111) are investigated across the hyperthermal energy regime (5 - 100 eV). Scattered ionic products are measured with mass-, angular-, and velocity-resolution. Scattered Br_2^-/Br^- product distributions are measured as a function of the incident Br_2^+/Br^+ collision energy, vibrational energy, and surface temperature. The yield for both atomic and molecular projectiles reveals an unusual behavior where negative ion conversion is most efficient for incident velocities near 7.6 km/sec. This sharp peak in the yield shifts to lower velocities with increasing surface temperature. A detailed analysis of the products' velocity distributions suggests an important interplay between charge transfer and energy transfer in this system.

2:40pm **SS1-WeA3 Hyperthermal Ejection of Atomic Cl from the Reaction of Cl_2 on the Al(111) Surface: Evidence of a Nonadiabatic Electron Harpooning Mechanism**, ***G.C. Poon***, *T.J. Grassman, A.C. Kummel*, University of California, San Diego

Resonantly Enhanced Multiphoton Ionization (REMPI) and Time-of-Flight Mass Spectroscopy (TOF-MS) have been used to demonstrate that the reaction of Cl_2 on the low work function Al(111) surface proceeds via a prompt nonadiabatic electron harpooning process. As Cl_2 approaches the Al(111) surface, an electron harpoons from the surface, suddenly converting Cl_2 to Cl_2^-. This places the molecule high on the repulsive portion of the Cl_2^- potential curve leading to rapid dissociation into Cl^- and Cl fragments. The Cl^- proceeds toward the surface and sticks while Cl is ejected into the gas phase. An experimentally observable signature of this harpooning process would be a hyperthermal translational energy of the ejected fragment, whose energy is determined by the vertical transition between Cl_2 and Cl_2^- and should be nearly independent of incident translational energy. Three beams of Cl_2 were prepared by seeding with translation energies ranging from 0.989 eV to 0.105 eV. Each of the three Cl_2 beams was directed at the surface at three incident angles: 0°, 20°, and 40°. The translational energy of the ejected Cl was shown to be a weak function of the incident translational energy. For 0.989 eV normal incidence Cl_2 the ejected Cl had a translational energy of 0.147 eV, while 0.105 eV normal incidence Cl_2 produced ejected Cl of 0.100 eV. Therefore, for incident Cl_2 with a velocity of 535 m/sec, the ejected Cl atoms were accelerated to a velocity of 740 m/sec. This acceleration of ejected Cl compared to incident Cl_2 is consistent with a nonadiabatic process that converts electronic to kinetic energy.

3:00pm **SS1-WeA4 Degradation of Alkanethiol Self Assembled Monolayers under Hyperthermal O^+ Bombardment**, ***T. Tzvetkov***, *X. Qin, D.C. Jacobs*, University of Notre Dame

We present experimental results on the reaction of 5-20eV O^+ with alkanethiol self-assembled monolayers (SAMs). Decanethiol or 1H, 1H, 2H, 2H-perfluorodecanethiol are used to form well-ordered SAMs on a clean gold surface. Scattered ionic products, formed as a result of hyperthermal O^+ ion bombardment, are collected with angle-, energy-, and mass-resolution. Chemical modifications in the SAM layer are monitored by X-ray photoelectron spectroscopy (XPS). Efficient removal of H from alkanethiol SAMs is observed at all incident energies. With increasing collision energy, C-C bond cleavage becomes important, as various scattered species of C_nH_m or C_nF_m radicals are observed, and the stoichiometry of the irradiated SAM is altered. The experimental results help us to understand the mechanisms of polymer degradation under O^+ bombardment at hyperthermal incident energies.

3:20pm **SS1-WeA5 Charge Transfer in Low Energy Li Ion Scattering from Halogen-covered Metal Surfaces**, ***Y. Yang***, *J.A. Yarmoff*, University of California, Riverside

Resonant charge transfer (RCT) for 400-3000 eV Li ions scattered from iodine- and bromine-covered Fe(110) and Ni(100) surfaces is probed with time-of-flight spectroscopy. RCT with alkali ions has traditionally been considered to reflect the overlap between the local surface potential and the ionization level of the exiting ion. In this work, it is shown that the local charge density can also influence the RCT process. For example, iodine would be expected to adsorb with some net negative charge, thereby producing a dipole directed into the surface that would raise the work function. In contrast, iodine decreases the work function, and the neutralization probabilities for Li singly scattered from the I sites are always higher than for scattering from metal sites. Thus, there is a local effect involved in RCT for this system. Bromine adsorption does not change the work function significantly, but still the neutralization probabilities for scattering from Br sites are larger than from the substrate. These results suggest that the local charge density does plays a significant role in determining the RCT probability.

3:40pm **SS1-WeA6 Adsorption and Radiation Induced Decomposition of SF_6 on Ru(0001)**, ***N.S. Faradzhev***, *D.O. Kusmierek, B.V. Yakshinskiy, T.E. Madey*, Rutgers, The State University of New Jersey

Electron stimulated desorption ion angular distribution (ESDIAD) and temperature programmed desorption (TPD) techniques have been employed to study the adsorption and radiation-induced decomposition of fractional monolayers of octahedrally-coordinated SF_6 physisorbed on Ru(0001) at 25K. TPD reveals predominantly molecular adsorption of SF_6, which desorbs from 3 weakly-bound states below 100K. ESDIAD of both F^+ and F^- ions at 25K demonstrate "halo-like" patterns, and heating the substrate to ~90K results in formation of hexagonal angular distributions for both ions. ESD of F^+ and F occurs via different mechanisms but from the same chemical states of molecular SF_6, which appears to be adsorbed via three F atoms, with the other three pointed away from the surface. At low temperatures, the F atoms have a random azimuthal orientation, while upon annealing, lateral ordering occurs in two complementary domains. Prolonged electron beam exposure leads to dissociation of SF_6, and formation of SF_x (x=0 to 5) fragments. F^- ions escape only from undissociated molecular SF_6, while F^+ ions also originate from dissociation fragments; both normal beams and off-normal beams are seen in hexagonal F^+ ESDIAD patterns, and intensities vary with electron exposure. Electron exposures $>10^{16}$ e/cm^2 result in complete decomposition of SF_6, as verified by TPD and ESDIAD.

4:00pm **SS1-WeA7 Dynamical Behaviors of GaCl on GaAs Surfaces by Pulsed Molecular Beam Scattering**, ***M. Ohashi***, National Institute of Advanced Industrial Science and Technology (AIST), Japan, *M. Ozeki*, Miyazaki University, Japan

Gallium chloride (GaCl) is an important precursor in the growth of compounds containing Ga and plays an important role in halide vapor-phase epitaxy (VPE) and hydride VPE. The GaAs layer grown on a GaAs(001) substrate is completely mirror-like without surface defects, but the layer grown on a GaAs(110) substrate is slightly hazy. The cause of these results is unclear. As the initial process in GaAs epitaxial growth is defined that GaCl molecule trapped into a precursor state in GaCl/GaAs surface system, this precursor plays an important role in growth. The reason that a high quality layer can be grown on a GaAs(001) substrate but not a GaAs(110) substrate has not clarified because the dynamical behaviors in the precursor states of GaCl on these surfces, such as activation energy and desorption rate have not been clarified. We investigated the adsorption mechanism of GaCl on the GaAs(001) and GaAs(110) surfaces based on angular distribution and the temperature dependence of GaCl time of flight spectra reflected from these surfaces. These well-defined surfaces are prepared by molecular beam epitaxy system connected with molecular beam scattering apparatus. The angular distribution of reflected GaCl consists of two parts; inelastic direct scattering contribution and thermal desorption of trapped molecules on the GaAs(001) and GaAs(110) surfaces. We divided the time of flight spectra of thermally desorbed GaCl into the component with activation energies of 92 kJ/mol corresponding to GaCl trapping well on GaAs(001) 2x4 surface and the components with 54 kJ/mol on GaAs(110) 1x1 surface Trapped GaCl desorbed rabidly from the GaAs(110) 1x1 surface compared to GaAs(001) 2x4 surface. These results suggest that the interaction between GaCl molecules and GaAs surface deeply influences the quality to large grown on GaAs substrates. Most of above this work was supported by New Energy and Industrial Technology Development Organization (NEDO).

4:20pm **SS1-WeA8 A New Mechanism for Ion-Stimulated Surface Processes at Low Energies**, *Z. Wang*, ***E.G. Seebauer***, University of Illinois

Ion surface interactions at low energies (<100 eV) characterize an increasingly diverse array of material processing steps in ion beam assisted deposition (IBAD), plasma enhanced deposition, reactive ion etching (RIE), and other applications. Overall process behavior in these applications often reflects a delicate balance among several competing kinetic effects. The governing kinetic phenomena are often tacitly considered to lie at one of two poles: physical effects where momentum matching dominates, and chemical effects involving thermal activation of atomic bonds according to Arrhenius expressions. Here we report molecular dynamics simulations of low-energy noble gas atoms impacting Si and Ge surfaces, and describe a new phenomenon that lies at neither pole. The simulations, backed by experiments on surface diffusion, exhibit a surprising new form of tradeoff between the ion energy threshold for point defect formation and substrate temperature. Because of the vast difference in scales between ion energies and thermal energies, the tradeoff resembles an elephant being balanced by a mouse on a seesaw. The effect originates from instantaneous nonuniformities in net surface potential induced by thermal vibrations, which dramatically affect the locality of momentum transfer to the surface and greatly amplify the effect of temperature. This amplification may offer a new means for selecting specific elementary rate processes during plasma processing or ion beam assisted deposition by judicious tuning of temperature and ion energy.

4:40pm **SS1-WeA9 Photochemistry of Caged Molecules: CD_3Cl in Ice**, ***M. Asscher***, *Y. Lilach*, Hebrew University of Jerusalem, Israel

The interaction of two similar coadsorbed dipolar molecules, H_2O and CD_3Cl, has been studied over Ru(001) under UHV conditions. The complementary techniques of δP-TPD and work function change in a $\delta\phi$-TPD mode were employed. Post-adsorption of water leads initially to compression, reorientation and then to the formation of CD_3Cl layers that are encapsulated between amorphous solid water layers. These caged molecules are explosively desorb at 165K. Unique photochemical reactivity is displayed by the trapped methyl chloride molecules upon irradiation by 6.4 eV photons from an ArF excimer laser. Radical chemistry based on hot, photochemically generated methyl fragments, lead to the formation of longer chain hydrocarbons as well as oxygenated products. The relevance of these observations to the origin of interstellar hydrocarbons is discussed.

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