

Photolysis and Radiolysis of Outer Solar System Ices (PROSSI) Workshop

Contributed Abstracts

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Bahr: Photodesorption and Photochemistry of Condensed Gases and Astronomical Implications

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ABSTRACT:

We will report absolute measurements of total photodesorption yields for condensed water, oxygen, carbon monoxide and carbon dioxide for irradiation with Lyman-alpha (10.2 eV) photons.

The absolute yields are determined with a highly sensitive microbalance technique, and relative desorption yields of photolysis products with a mass spectrometer. We will discuss the implications of photolysis and photodesorption in the surface chemistry of icy satellites and interstellar grains.

Bahr: Brightening of Amorphous Solid Water by Cracking and by Ion Irradiation: Implications for Optical Remote Sensing of Icy Satellites

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ABSTRACT:

Using optical microscopy, we have studied the surface of films of amorphous solid water grown by vapor deposition in ultrahigh vacuum. We found that cracks form when the thickness of the ice films exceed a critical value, and we follow the evolution of the cracked surface with further deposition of water. The geometry of the cracks and the critical thickness depend on deposition temperature. We will discuss these findings in terms of porosity of the ice films and stresses during film growth. Since cracking produces strong brightening it needs to be considered when modeling the optical reflectance of icy satellites. We will discuss brightening resulting from ion irradiation based on new laboratory simulations.

Baragiola: Ozone Synthesis at Ganymede

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ABSTRACT:

We will present new measurements on ozone synthesis in condensed, oxygen-bearing gases by 100 keV proton impact. Ozone is detected by reflectance spectroscopy in the ultraviolet (Hartley band) and by mass spectrometry during thermal desorption. We will compare band shapes obtained in the laboratory with those resulting from observations by UV spectrometers on the Hubble Space Telescope and Galileo space probe. We will also report model calculations of ozone chemistry in Ganymede which takes into account both the irradiation by ions and by solar photons.

Baragiola: Sputter Production of Water and Oxygen Atmospheres at Icy Satellites: Temperature Effects

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ABSTRACT:

Energetic photons, electrons and ions erode the surface satellites ejecting molecule to the gas phase, where they contribute to the formation of atmospheres. We will present measurements on yields of sputtering (desorption) of water and oxygen molecules from solid water induced by 5-100 keV ions and Lyman-alpha photons, in the temperature range between 10 K and 160 K. We will discuss physical mechanisms and implications of the results for the formation of atmospheres around the icy satellites of Jupiter and Saturn.

Carlson: Abundance of sulfur, sulfur dioxide, sulfuric acid, hydrogen peroxide, and carbon dioxide on Europa's surface and their radiolytic production and destruction processes.

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ABSTRACT:

Europa observations and laboratory optical data are used to estimate molecular abundances in Europa's optical surface layer, which is a fraction of a mm to a few mm thick, depending upon the wavelength. The average concentration of sulfur on the trailing side is estimated to be $[S]/[H_2O] \sim 0.06\%$ to 1.2% for mixtures of sulfur polymers in ice. An ultraviolet-derived SO_2 average abundance for the trailing side is $[SO_2]/[H_2O] \sim 0.03\%$, while an infrared-based leading side abundance is approximately the same, but subject to large observational and experimental uncertainties. Hydrated sulfuric acid ($H_2SO_4 \cdot nH_2O$, $n = 6.5, 8$) is the dominant species in portions of the trailing hemisphere, where $[H_2SO_4]/[H_2O] \sim 10\%$. The corresponding global average is a few percent. On the leading side, $[H_2O_2]/[H_2O] \sim 0.1\%$ while $[CO_2]/[H_2O] \sim 0.01\%$. Upper limits for NaOH and $Mg(OH)_2$ are 5% and 3% respectively and interesting because one suggested source of Europa's sulfur is internally derived Na and Mg sulfates.

Radiolytic production and destruction processes are evaluated for the above species using Cooper et al.'s Europa irradiation fluxes with Venus and terrestrial sulfur photochemistry and laboratory irradiation experiments. Cross sections and efficiencies (commonly expressed as G-values, the number of molecules produced or destroyed per 100 eV of energy absorbed) are available for some processes, but data for Europa temperature conditions are needed. Oxidation of elemental sulfur to form the H_2SO_4 end product proceeds through reactions with radiolytically produced O, O_2 , O_2^- , H_2O_2 , OH, and HO_2 , forming SO, $S(OH)_2$, SO_2 , SO_3 , and others as transient intermediates. Once produced, hydrated H_2SO_4 is relatively stable under irradiation ($G < 0.001$) due to regeneration that occurs in back reactions. This relatively slow destruction, producing SO_2 and S_x in the continuous radiolytic sulfur cycle, results in the observed large abundance of H_2SO_4 hydrate. Magnesium hydroxide is also quite stable ($G < 0.03$), so the above limit indicates a low rate of $Mg(OH)_2$ production, implying either low Mg concentrations or low production efficiencies.

Cornelison: X-Ray Induced Percolation Transition in the Diffusion of Photo-Dissociated Species in Solid CO₂

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ABSTRACT:

Solid CO₂ films were grown at 77K and then photo-processed by x-rays. The species produced were monitored in the gas phase, using a quadrupole mass spectrometer. In earlier work, it was shown that several species are produced by x-ray photo-dissociation and that these products show anomalous behavior in their diffusion characteristics. Specifically, no diffusion is seen (as evidenced by a lack of gas-phase increases) for some characteristic time, after which a sudden onset occurs. The behavior of this characteristic time is studied as a function of x-ray power, and the system is described using a percolation model, which can explain the qualitative behavior of the gas-phase concentrations and yield a critical value for the disorder at which the transition takes place. Additionally, a model based on bulk diffusion and surface processes (reaction and desorption) has been developed for the CO₂ system. Using two free (diffusion coefficient and desorption probability) and two calculated (hydrogenation and pumping rates) parameters, the model is compared to the time dependent gas-phase concentrations for the important species of the system.

Cottin: Experimental and Theoretical Studies on the gas/solid/gas Transformation Cycle in Extraterrestrial Environments

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ABSTRACT:

Experimental and theoretical studies are performed in our laboratory in order to study, on one hand, the evolution from gases to solid material (S.C.O.O.P. program), and, on the other hand, the degradation of high molecular weight compounds into gas phase molecules (S.E.M.A.Ph.Or.E.Cometaire program). These programs provide spectroscopic and quantitative physicochemical data necessary to better understand photochemical and thermal processes involved in the gas/solid/gas transformation cycle that occurs in environments such as interstellar medium, planetary and cometary atmospheres.

Cyanopolyynes ($\text{HC}_{(2n+1)}\text{N}$) and very recently polyynes (C_{2n}H_2) have been observed in the interstellar medium. Dicyanopolyynes (C_{2n}N_2) are certainly also present. Those compounds are key molecules in the formation of higher molecular weight compounds (like PAHs) which contribute to grain formation in ISM. HC_3N , C_4H_2 , C_4N_2 have been observed in planetary atmospheres where the photochemistry of methane (and nitrogen) leads to the formation of solid particles (like tholins). Spectroscopic data of those long carbon chains are needed to interpret observations and develop photochemical models. The aim of the S.C.O.O.P. program is to provide such crucial data at any temperature relevant to the studied environments. We will present absolute absorption spectra of polyynes (C_{2n}H_2 , $n=1,4$), HC_3N and C_4N_2 , obtained in the infrared and ultraviolet wavelength range.

Additionally to the experimental approach, a theoretical work is underway to

extrapolate the results to any temperature, and to longer carbon chains compounds susceptible to condense. At last, signatures of such molecules will be known and their quantification allowed. At their time, through different processes, as thermal- and photo-degradation, high molecular weight molecules in solid phase can release volatile products, which may be different than their original precursors, and thus act as parent molecules for extended sources in comets, and indirect pathways for the production of interstellar species. The aim of the S.E.M.A.Ph.Or.E. Cometaire program is to bring a better understanding of the chemical mechanisms involved during those transformations, and provide physico-chemical data such as the quantum yield of production of gaseous molecules from the solid parent molecule as a function of the wavelength of UV irradiation. We have shown, for example, that photodegradation of polyoxymethylene [P.O.M. : $(-\text{CH}_2\text{-O}-)_n$] in the 122-193 nm range produce daughter gaseous molecules like H_2CO , CO , CO_2 , HCOOH , CH_3OH , CH_3OCHO , already detected in comets, $\text{CH}_3\text{OCH}_2\text{OCH}_3$ and $\text{C}_3\text{H}_6\text{O}_3$ (trioxane). We have derived production quantum yield as a function of the wavelength for some of them. Such data were missing to interpret and model observations. Now, they allow us to answer in what extend the presence of P.O.M. is relevant as an explanation for the formaldehyde extended sources observed in several comets. Study of other molecules is in progress or planned: hexamethylenetetramine ($\text{C}_6\text{H}_{12}\text{N}_4$), HCN polymer, tholins, PAHs, and amino acids. Interaction between those molecules and water photofragments may also an interesting mechanism to investigate.

In spite of the fact that the aim of both programs is to describe with the better accuracy two reverse phenomena (evolution of gases to solid and degradation of solid to gases), the methodological approach is the same. It consists, in both case, to proceed step by step with an increasing complexity of the studied compounds or mechanisms. In our point of view, performing such studies will allow to clarify many aspects of the highly complex chemical evolution of extraterrestrial environment.

Delitsky: Organic Chemistry and Phase Effects on the Galilean and Saturnian Satellites

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Outer planet satellites are subjected to irradiation from magnetospheric ions, photons, solar wind particles and cosmic rays. Ions will be implanted in their surfaces initiating chemistry at these sites, which is affected by factors such as heating and cooling from day-night cycles, interstitial disequilibrium, local temperature (latitude), and the oscillation of the magnetosphere.

Since in Ganymede's own magnetosphere, the field lines direct ions into its poles, disruption of crystal structures there should make the water ice amorphous. Phase effects should influence the local chemistry. Also, water radiolysis products, such as ozone, will be below their melting points at the poles (80 K for O₃), and should be liquid in the equatorial regions, so spectral absorbances will be different in polar versus equatorial regions.

The presence of CO₂ on Ganymede and Callisto, as detected by the Galileo spacecraft, implies the presence of a whole host of organic compounds, as suggested by Delitsky and Lane (1998), including CH₃OH, HCOOH, CO, HCO, CH₃CH₂OH, (CH₃)₂CO, HOCH₂CH₂OH, CH₃COOH, CO₂, H₂CO₃, H₂CO, C₃O₂, CH₂CO, and others. On the Saturnian satellites, nitrogen ions from sputtering of Titan's atmosphere may impact the surfaces of the other satellites, Rhea, Dione, Tethys, Enceladus, (if the ions can migrate inwards towards Saturn) and form such species as NO₂, HNO₂, HNO₃, NH₂OH, and R-OCN (Strazzulla 1999, Roessler 1985,1986).

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- 2) Roessler, Cosmic chemistry of carbon and nitrogen implants in ice, Origins and Early Evolution of Life (Proceedings), Terra Cognita, 5,128(1985)
- 3) Roessler, Chemical modification of insulators by ion-implantation, Rad. Effects and Defects in Solids, 99, 21(1986)
- 4) Strazzulla,1999, Ion irradiation experiments and nitrogen bearing species on Jovian and Saturnian icy surfaces, Planetary and Space Sci 47,1371(1999)

Delzeit: Structural Characterization of Crystalline Ice Nanoclusters

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ABSTRACT:

Water ice nanoclusters are useful analogs for studying a variety of processes that occur within icy grains in the extraterrestrial environment. The surface of ice nanoclusters prepared in the laboratory is similar to the surface of interstellar ice grains. In cold molecular clouds, the silicate cores of interstellar grains are typically ~100 nm in diameter and have a coating of impure amorphous water ice. Depositional, thermal and radiolytic processes leave the surface and subsurface molecules in a disordered state. In this state, structural defects become mobile and reactions of trapped gases and small molecules can occur. The large surface area of nanocluster deposits relative to their bulk allows for routine observation of such surface-mediated processes. Furthermore, the disordered surface and subsurface layers in nanocluster deposits mimic the structure of amorphous ice rinds found on interstellar dust grains.

Transmission Electron Microscopy (TEM) has been used to characterize the crystallinity, growth mechanism, and size distribution of nanoclusters formed from a mixture of water vapor with an inert carrier gas that has been rapidly cooled to 77K. TEM imaging reveals a Gaussian size distribution around a modal diameter that increases from approx. 15 to 30 nm as the percentage of water vapor within the mixture increases from 0.5 to 2.0%, respectively.

TEM bright and dark field imaging also reveals the crystalline nature of the clusters. Many of the clusters show a mosaic structure in which crystalline domains originate at the center of the cluster and grow radially outward. Other images show mirror planes that are separated by approx. 10 nm. Electron diffraction patterns of these clusters show that the clusters are composed of cubic ice with only a small hexagonal component. Further, the crystalline domain size is approximately the same as the modal diameter suggesting that the clusters are single crystals.

d'Hendecourt: The chemical composition of silicate dust around evolved stars and protostars

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ABSTRACT:

Although this meeting is entirely devoted to chemical evolution of ices upon bombardment of energetic particles (UV photons and/or cosmic-rays particles), one must not forget that these particles may severely affect the "core" of the grains, i.e. the silicates, largely observed in the Interstellar Medium. We present in this poster an analysis of the chemical composition of silicate dust around various objects, namely evolved stars and protostars.

Differences between these two lines of sight are obtained not only in the *physical structure* of the silicates (large proportions of crystalline silicates are present in evolved stars whereas those observed in protostars are totally amorphous but also in the *chemical nature* of the silicates (olivine in evolved stars and orthopyroxene and aluminosilicates in protostars). This poster describes in detail ISO spectra from these objects. Since the observed silicates relate to each other following a certain pattern of evolution of the grains, we postulate that most observed differences may be the result of fast particles bombardment of interstellar grains that modify their structure and chemical composition. Experiments at IAS are undertaken to test this hypothesis in order to retrace with quantitative arguments, the life of a grain in the ISM.

Dillingham: Surface Characterization and X-Ray Photolysis of Carbon Dioxide/Methanol, Carbon Dioxide/Ammonia and Water/Ammonia Ices

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ABSTRACT:

The investigation of ices and the photochemical processes that can occur in these ices have important applications in astrophysics, planetary astronomy and atmospheric physics. In this study, results are presented concerning carbon dioxide/methanol, carbon dioxide/ammonia and water/ammonia ices formed in an ultra-high vacuum environment (base pressure 5×10^{-10} Torr) at liquid nitrogen temperatures. The ices are characterized using x-ray photoelectron spectroscopy (XPS). In the surface sensitive technique of XPS, the ice to be studied is irradiated with x-rays and the energies of the ejected photoelectrons are measured. A precise measurement of the energies of these electrons affords a good means of elemental identification. Furthermore, since the photoelectron energy is dependent upon the local chemical environment of the atom, it is possible to distinguish particular details concerning the chemical state. Typically, the analysis depth, which corresponds to the mean free path of the photoelectron, is of the order of 10-20 Å. The chemical changes are monitored while the ices are continuously irradiated with Mg K α x-rays (photon energy 1253.6 eV and a flux of approximately 6×10^{13} photons/cm 2 -s) for periods of up to 5 hours.

For the CO $_2$ /CH $_3$ OH ice, the production of CO and H $_2$ CO is clearly observed following x-ray irradiation. Furthermore, significant changes are observed in the XPS core-level line shapes for the CO $_2$ /NH $_3$ ice while only minor changes are observed for the H $_2$ O/NH $_3$ ice. A quadrupole mass spectrometer was also used to monitor the species evolving from the ice surfaces during the photoprocessing. The mass spectrometer results confirm the production of CO and H $_2$ CO for the CO $_2$ /CH $_3$ OH ice. For the CO $_2$ /NH $_3$ ice, several species (especially carbon bearing molecules) are evolved from the ice surface while for the H $_2$ O/NH $_3$

ice, no significant changes are observed in the RGA plots. This last finding is consistent with the XPS results. The XPS and mass spectrometer results are thus correlated and possible mechanisms associated with the photochemical processing are discussed.

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Gerakines: Energetic Processing Laboratory Ice Analogs: UV Photolysis vs. Ion Bombardment

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ABSTRACT:

Two of the most important energetic processes thought to drive interstellar and Solar-System ice chemistries (apart from purely thermal effects) are ultraviolet (UV) photolysis and exposure to cosmic rays. Generally, these processes are considered equivalent, but few quantitative comparisons of UV- and ion-driven chemistries have ever been published.

The cosmic-ice laboratory at NASA/GSFC is capable of performing both UV photolysis and proton bombardment in the same experimental set-up, with samples created under the same conditions. Here we present direct quantitative comparisons of these effects on various mixtures of H₂O, CH₄, CH₃OH and CO₂ at T=18K. Equivalent energy doses have been applied in the form of UV photons (approx. 10eV/photon) or 0.8MeV protons. Product formation rates are compared and differences discussed in terms of the radiation/ice interaction in each case. Ramifications are discussed for ices in various Solar-System environments.

P.A.G. holds a National Research Council-NASA/GSFC Research Associateship.

Grundy: Peculiar Near-IR Spectral Features of Icy Satellites

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ABSTRACT:

We have been spectroscopically surveying icy satellites at near-infrared wavelengths since 1995. Our observations have made use of ground-based instrumentation at Lowell Observatory and Kitt Peak, as well as the NICMOS grism mode of HST. Whenever possible, we have observed targets repeatedly, in order to check consistency and to seek changes depending on varying illumination and observing geometry.

In our wavelength range (1.2-2.6 microns) the spectra of icy satellites are dominated by vibrational absorptions of H₂O ice, most notably at 1.5 and 2.0 microns, with weaker features at 1.3 and 1.65 microns. We made use of recent laboratory work providing temperature-dependent absorption coefficients of H₂O ice at high precision (Grundy and Schmitt 1998) in order to determine surface temperatures of icy satellites from their near-infrared spectra (Grundy et al. 1999, Buie and Grundy 2000).

During our analysis, we confirmed that some, but not all icy satellites exhibit spectral features apparently inconsistent with models based on the absorption coefficients of ordinary H₂O ice Ih and spectrally neutral continuum absorbers. Some of these features have been noted earlier (e.g., Clark et al. 1984, Calvin et al. 1995, McCord et al. 1998). We will concentrate on an anomalously deep 1.65 micron band (Enceladus), an albedo fall off at wavelengths long-ward of 2 microns (most Saturnian satellites and Charon), and peculiarities in the shape of the continuum region at 2.2 microns (some Saturnian and Uranian satellites and Charon). We will show examples of these not-yet-understood spectral features and will discuss their distribution in terms of heliocentric distance, leading versus trailing hemisphere, albedo, etc.

Acknowledgments:

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Grundy: Solar Gardening of Volatile Ices on Pluto and Triton

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ABSTRACT:

To accurately model the spectral reflectance of an icy surface, it is necessary to simulate radiative transfer within that surface. As a byproduct of this effort, one obtains the distribution with depth of absorbed sunlight. A similar radiative transfer calculation can be done to obtain the distribution with depth of the source of directionally emitted thermal radiation. By integrating over emission angle and wavelength, one can obtain bolometric emission as a function of depth. These emission and absorption distributions can be combined to yield the time- and depth-dependent description of energy sources and sinks within a planetary surface.

For many ice species, absorption coefficients are smaller at visible wavelengths than at infrared wavelengths. Solar energy is thus, on average, absorbed more deeply within the surface than the depth from which thermal energy is, on average, emitted. Differences between the two vertical distributions require energy transport from where the energy is absorbed to where it is emitted. For surfaces composed of non-volatile materials on airless bodies, this transport is achieved by the establishment of a thermal gradient, or "solid state greenhouse" which has been explored in some detail elsewhere.

In the case of the nitrogen ice complexes of Pluto and Triton, which are in vapor pressure equilibrium with nitrogen-dominated atmospheres, energy is more efficiently transported by bulk flow of material. Examining this global time- and depth-dependent subsurface material flow, we find that the uppermost few grain diameters of bright, N₂ ice regions see a net deposition of material, even under conditions of maximum insolation, such as high southern latitudes on Triton during maximum southern summer. However, immediately below this uppermost layer, there is a larger region down to some tens of grain diameters, which experiences a net loss of material. These two trends result in a systematic overturning and recycling of the material in bright N₂ frosts with time scales of order years to decades, when they are illuminated by sunlight.

This "solar gardening" process has important repercussions for the surface texture of the ice, and thus for its photometric behavior. The picture is further complicated by solid state distillation, whereby the most volatile materials (e.g. N_2 and CO) sublime preferentially, leaving elevated concentrations of less volatile species (e.g. CH_4 and higher hydrocarbons). Ice in regions having experienced extensive sublimation will be enriched in less volatile species compared with more recently deposited ice, which has a composition similar to that of the atmosphere. We thus expect to see a layered structure in areas receiving strong insolation, with hydrocarbon-enriched ice beneath more depleted ice. Such a configuration would exhibit enhancement of weak hydrocarbon absorption bands relative to those of stronger bands, because photons at more weakly absorbing wavelength penetrate deeper into the surface.

Acknowledgments

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Hanlon: Optical Absorption by Ice

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ABSTRACT:

The aim of this project is to accurately determine the absorption spectrum (and hence the reflectance spectrum) of pure H₂O ice between 190 and 700 nm. The reflectance spectrum would provide a useful baseline for reflectance studies of ice on outer solar system bodies. Although some previous work [1-6] has been carried out in the UV and visible wavelength range by a few previous workers, the published absorption spectra often vary by more than an order of magnitude from one worker to another. There are a number of possible reasons for the differences between the absorption spectra. Firstly, water purification is of critical importance. Many workers simply used "distilled" water and do not include any steps designed to remove organic impurities which may absorb in the UV region of the spectrum. Furthermore, the large vertical displacements in the spectra are often due to light scattering by dislocations and crystal boundaries in the ice, as well as particulates. The absorption then appears to be higher, as some of the light is scattered out of the field of view of the detector.

In the present work special care is being taken to purify the water, by triple distillation incorporating an oxidative purification step to remove organic impurities. This water also has a very low particulate concentration. Large, single crystals of ice, approximately 1 meter in length, are produced by slowly lowering (at *ca.* 1 cm/hour) a tube of degassed purified water into a cold bath at -5° C to produce the initial ice sample. This sample is then further purified by the method of zone refining [7], to produce an optically clear single crystal ice sample. It is hoped that in this way, a definitive absorption spectrum (and the corresponding reflectance spectrum) will be obtained for ice between 190 and 700 nm, and that the large order of magnitude discrepancies from one worker to another will be removed.

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Hansen: Amorphous And Crystalline Ice On The Galilean Satellites: A Balance Between Thermal And Radiolytic Processes.

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ABSTRACT:

Water ice is abundant on the three outermost Galilean Satellites of Jupiter, Europa, Ganymede, and Callisto [1]. We use near-infrared spectra returned by the Near-Infrared Mapping Spectrometer (NIMS) on the Galileo spacecraft to study the crystalline order of the surface ice on all three satellites. The lattice order of ice is known to be dependent on its condensation temperature and rate, and its temperature history [2]. The surface temperature of these satellites is in a range where any amorphous surface ice will crystallize over very short time scales [3]. Based on this, we would expect to see predominantly well crystalline ice on the outer satellites and perhaps less crystalline ice on Europa which has the coldest (and youngest) surface. Ice in the laboratory will crystallize almost instantly at 140-150 K, while the ice temperatures on the outer satellites is probably in the 120-130 K range [4]. Kinetic models and studies of ice indicate that crystallization at this temperature will occur within about 1 yr. [3].

European ice is 10-20 K colder and will stay amorphous for a somewhat longer time. New amorphous ice can be created from vapor deposition or flash freezing of cryovolcanic liquids. A more likely source of new amorphous ice is the disruption of crystalline ice by corpuscular radiation, which is strong and widespread throughout the Galilean satellites, since radiation is known to cause disorder in crystalline ice [5]. We can envision a balance between thermal kinetic crystallization and radiolytic disruption, where cold temperatures and high radiation fluxes would favor amorphous ice on Europa, while much lower radiolytic fluxes and higher temperatures on Callisto would favor crystalline ice, with Ganymede falling in between. The intrinsic magnetic field of Ganymede diverts much of the local plasma flux into the higher latitudes. This fact has been used to understand the polar brightening seen on Ganymede as a locally derived (amorphous?) frost from sputtering by the high polar radiation levels [6].

There are several features in the near-infrared reflection spectrum of water ice which can be used to probe lattice order. This includes the temperature-sensitive band at 1.65 microns, which disappears altogether in cold amorphous ice [7], and the fundamental O-H-stretch absorption near 3.1 microns, which appears as a reflection peak, and is stronger and has higher spectral detail for crystalline ice [8]. The 1.65-micron arises from ~1 mm below the surface, while the 3.1-micron peak comes from the Fresnel reflection off the facets of the water ice grains in the surface, and so is effectively from zero depth. Our analysis of the 3.1-micron feature in NIMS spectra shows that the zero-depth ice is crystalline on Ganymede and Callisto, and disordered on Europa. The 3.1-micron peak on Ganymede appears to vary over the surface, and is most non-crystalline in the polar regions, supporting the theories of higher radiation fluxes there. The exact position and width of the band in amorphous ice is dependent on temperature and degree of order [8]. We will compare the implied widths and centers of the features on Europa and Ganymede to laboratory studies. The 1.65-micron band appears everywhere, implying that the amorphous layer is very thin.

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Hendrix: Galileo UVS Spectra of the Icy Galilean Satellites: Evidence for Photolysis and/or Radiolysis

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ABSTRACT:

We report on Galileo Ultraviolet Spectrometer (UVS) data of the icy Galilean satellites Europa, Ganymede and Callisto. Results are compiled from three years of observations, spanning the Galileo primary and extended missions. Our results focus on the detection of minor species involved in chemistry of the surface ice. The source of the ice chemistry may be solar photons, charged particles, or both. Species that have been detected are hydrogen peroxide, ozone (as determined by an absorption band near 260 nm), and a sulfate-type species (as determined by an absorption band near 280 nm); escaping hydrogen atoms have been detected at Ganymede and Callisto.

Europa exhibits the strongest hydrogen peroxide feature of the three icy satellites, consistent with its relatively pure ice surface. Europa also displays an absorption band near 280 nm that appears to be associated with dark terrain, primarily on the trailing hemisphere; the band may be due to sulfates of some sort and, if due to dark terrain, is likely endogenic in nature. On Ganymede, hydrogen peroxide varies clearly with solar angle, where more hydrogen peroxide exists at small solar angles. This is in contrast with ozone, which generally exists in larger quantities at larger solar angles. Observations of Callisto show that hydrogen peroxide amounts increase with decreasing solar angle while the ozone-like absorber and a sulfate-type absorber are present in distinct regions. In particular, variations are noted between the hemispheres containing the large multi-ringed basins Asgard and Valhalla.

Khriachtchev: Photochemistry of hydrogen peroxide in rare-gas solids

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ABSTRACT:

UV photolysis of hydrogen peroxide (H_2O_2) in various rare-gas matrixes (Ar, Kr, and Xe) is comparatively studied. The photorecovery of H_2O_2 from the tight $\text{H}_2\text{O}\cdots\text{O}$ complex is observed in all rare-gas matrixes. The similarity of spectral position and efficiency of the photorecovery reaction in various rare-gas solids indicates its fundamental character, supports charge-transfer excitation of $\text{H}_2\text{O}\cdots\text{O}$ as its origin, and preserves promises to find this photoreaction in media of environmental importance, particularly in the gas phase and water ices. Our experiments on 193-nm photolysis of water dimers in solid Xe strongly support this hypothesis showing photogeneration of monomeric H_2O_2 .

**Lowenthal: Infrared Investigations of Relevant Planetary and Interstellar Ices:
Chemistry of Formation of Ammonium Cyanate/Ammonium Carbonate**

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ABSTRACT:

Proton irradiation of mixed ices of NH_3 , CO_2 , and H_2O at low temperatures ($\sim 10\text{K}$) are being investigated using FT-IR spectroscopy as the principle technique. Preliminary studies provide evidence for the formation of cyanates from proton bombardment of these ices. The observed 4.6 micron band in several interstellar spectra has tentatively been assigned to the cyanate ion (NCO^-) while the observed 6.8 micron band appears to have contribution from both $\text{CO}_3=$ and NH_4^+ ions. Results from the studies of the kinetic conversion of aqueous NH_4NCO to NH_4CO_3 are presented. The cyanate ion undergoes a quantitative conversion to $\text{CO}_3=$ and NH_4^+ in aqueous solution. Possible pathways for this conversion are under investigation. Aqueous ammonium cyanate (NH_4NCO) cooled to $\sim 10\text{K}$ exhibits the relevant 6.8 and 4.6 micron peaks in the infrared region that are observed in interstellar spectra. The results of this laboratory work are being utilized to examine the possible pathways for the production of NCO^- , $\text{CO}_3=$, and NH_4^+ ions in interstellar grains/ices. These studies may also be of relevance to cometary spectra.

Mastrapa: Trapping of Volatile Gases in Amorphous Water Ice

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ABSTRACT:

Introduction: Objects from the Kuiper Belt are prime candidates for the study of photolysis and radiolysis of icy bodies. It is assumed that Kuiper Belt Objects (KBOs) have remained at low temperatures at the outer edge of the Solar System since formation. Before determining what long term effects photolysis and radiolysis have had on these objects, one must first attempt to determine the composition of these objects, i.e. what chemical components are available for processing. To that end, this project examined the amount of volatiles trapped in amorphous water at low temperatures as an analog to formation of KBO's.

Experimental Procedure: All experiments were conducted at the Extraterrestrial Ice Simulator (EIS) at JPL. CO, CH₄, and CO₂ were trapped in H₂O at temperatures ranging from 20 K to 100 K and pressures between 10⁻⁵ and 10⁻⁸ Torr (~10⁻⁸ - ~10⁻¹¹ bars). The gases were released into the sample where they condensed on a sapphire window. This window is connected to a Helium refrigerator, and is therefore the coldest place in the chamber. Each experiment involved two gases at a time, water and either CO, CH₄, or CO₂. An infrared transmission spectrum was taken of each sample before sublimation to confirm the presence of water and the other volatile. The samples were then heated at rates between 0.25 K/min and 1 K/min. During heating, a mass spectrometer was used to measure the amount of volatiles escaping from the chamber. This flux was then integrated to get the total number of molecules condensed on the window.

Results and Conclusions: The volatiles escaped from the ice mixtures in three temperature ranges: 48-52 K, 145-160 K, 170-185 K. These ranges are similar to those found in previous work [1, 2, 3, 4]. In this experimental set up, water sublimates from 150 K to 185

K. However, the temperature range of escape is strongly dependent on the deposition temperature. If the deposition temperature is below the point where the solid volatile rapidly sublimates in the ambient environment of our experiment, from now on referred to as the solid point, then the first range of volatile escape is centered around its solid point, and there is little of the volatile remaining from 170-185 K. At the ambient pressure of the sample chamber, the solid points of CH₄, CO, and CO₂ are, respectively, 45 K, 40 K, and 80 K. Our results show that the ratio of water to volatile increases with temperature of deposition. However there is a significant number of volatile molecules still present at higher deposition temperatures. Volatiles such as CH₄ and CO can be trapped in amorphous ice at temperatures above their solid point. Future experiments will include measurements with different volatiles, such as O₂, and Ar. Also, since the volatiles were released into the sample chamber at the same pressure as the water, measurements will be taken with a solar system composition gas with the proper ratios of water to volatile. We also plan to introduce more than two gases at a time into the system. Once we have determined the amount of volatiles that can be trapped, we plan to conduct experiments to determine the effect of photolysis on trapped gases.

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Paranicas: The Radiation Environment of Icy Solar System Objects

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ABSTRACT:

Icy planetary satellites, short-period comets, and icy Kuiper belt objects are continuously bombarded with plasma and energetic charged particles. Flux rates to the surfaces of these objects vary considerably for a number of reasons. Most important is the influence of the local environment. Objects in the outer heliosphere are subject to the tenuous solar wind plasma, galactic and anomalous cosmic rays and variable fluxes of energetic particles associated, for instance, with solar proton events and shocks or “shock remnants.” On the other hand, satellites in magnetospheres are typically shielded from the solar wind environment. These bodies may instead be exposed to a highly variable distribution of trapped particles associated with the planet’s radiation belts and plasma sheet. Other aspects of the ambient plasma such as composition and charge state also play a role. The Jovian satellites are typically bombarded by a plasma rich in heavy ions.

In addition to environment, there are the properties of the body itself. Icy surfaces can be effectively shielded from a whole class of charged particles on account of the electromagnetic properties of the body, its atmosphere, ionosphere or magnetosphere. For instance, the Ganymede magnetosphere stands off the corotating Jovian plasma high above its surface; energetic particles have much greater access to polar caps than equatorial latitudes. Other satellite electromagnetics are not as straightforward to understand. For instance, Europa’s induced magnetic field and “Alfven wing” effects make it difficult to describe magnetic field topology near that object’s surface. These considerations are important for assessing whether observed features, such as leading/trailing asymmetries inferred by Galileo UVS measurements, can be correlated with variations in surface access by the radiation environment.

In this talk we will review the plasma and energetic charged particle data relevant to icy surfaces in the outer solar system. In addition, we will review work which connects these measurements to surface processes such as chemical modifications and sputtering. Finally, for icy object environments which have not been studied *in situ*, we will make predictions about surface modifications based on solar wind data.

Prockter: Deconvolving Exogenic and Endogenic Processes on Ganymede and Europa

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ABSTRACT:

The surfaces of icy satellites are affected by both endogenic (internal) and exogenic (external) processes, and deconvolving the effects of these processes on surface albedo variations can be difficult. Ganymede, the largest moon in the Solar System, has a surface distinctly divided into dark terrain (old, low albedo, heavily cratered terrain), and bright terrain (younger, high albedo, heavily ridged terrain). Bright terrain appears to be predominantly composed of water ice, while dark terrain is largely water ice with some percentage of a low albedo silicate contaminant mixed in [1]. Europa has a water and/or ice shell ~100 km thick [2], with a variety of reddish lineaments and disrupted terrain distributed across its surface. These features are contaminated by hydrated materials such as salts and/or sulfuric acid, potentially derived from a subsurface ocean [3, 4]. High-resolution Galileo imaging reveals distinctive albedo patterns on each of these satellites. While some processes causing albedo variations on these moons are relatively well-understood, there are several outstanding questions regarding the way in which their surfaces have evolved as a result of exogenic processes over time. Here we review the current understanding of the distribution of bright and dark materials with different albedos on these two moons, and highlight outstanding questions regarding their origins.

Ganymede

High resolution Galileo imaging has revealed a surface that is segregated into bright and dark patches, indicating that thermal segregation likely operates on this moon at small scales [5, 6]. These observations have led to a model in which the subsurface of Ganymede consists of a dirty ice layer (ice contaminated with dark meteoritic material) overlying cleaner ice [7]. Over time, solar radiation removes ice from the relatively warm dark terrain and deposits it in colder areas as frost, at the same time leaving dark material concentrated on the surface as a low albedo lag deposit. Bright terrain is proposed to form by the tectonic disruption of older dark terrain, slicing it into large tilted fault blocks, potentially in combination with icy volcanism [8]. As tilted blocks form, the dark veneer from their surfaces is proposed to slough into the valleys between the blocks, increasing the integrated albedo. In polar regions the dominant process affecting the surface is likely sputtering by energetic particles, apparently channeled via open magnetic field lines onto the surface [9]. Cold trapping and thermal segregation of sputtered ice also appears to operate at these high latitudes, as some thick bright deposits are observed [10], although radiation damage may also affect ice albedo [11]. The relationship between radiation damage and thermal segregation on frost deposition at the polar caps is an important outstanding question.

Europa

Although largely composed of water ice, Europa's shell is scarred by dark lineaments, and regions of disrupted terrain [12]. The lineaments, proposed to form along cracks created by secular and diurnal tidal stressing, are commonly reddish in color, and are proposed to contain traces of hydrated salts and/or sulfuric acid [3,4]. Regions of disrupted terrain, known as mottled terrain [e.g., 12], show the same signatures, prompting suggestions of a global reservoir of material [3]. At near infrared wavelengths (0.968 μm), the European ridged plains appear relatively dark in Galileo images, presumably as a result of the high percentage of coarse-grained water ice [13]. Europa also has a yellowish tinge on its leading hemisphere, potentially the result of staining by sulfur from Io [14]. At visible wavelengths, many of the wider lineaments, or bands, appear dark when relatively young, but become gray with time, eventually fading to the same high brightness as the background plains. Originally, this was suggested to result from frost deposition [15], but at high latitudes, infrared observations show that these lineaments become brighter than the background plains [13], suggesting that some other process has operated.

Outstanding questions for Europa include: (1) What is the relative rate of thermal redistribution vs. sputter redistribution? (2) Does endogenic material on Europa's surface begin dark or bright, and how quickly does its albedo change? (3) What are the relative roles of sputtering, thermal segregation, radiation damage, and chemical change in the brightening and spectral changes of ice, and can we estimate timescales for these factors?

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Robinson: FTIR-RAS and vapor pressure characterization of low density amorphous, restrained amorphous and crystalline ice during annealing between 90-170 K

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ABSTRACT:

Experimental evidence is provided for three low-temperature, metastable structures of water ice characterized as low-density amorphous, restrained amorphous and cubic crystalline [1]. Ice films are vapor deposited at 90 K in a high vacuum stainless steel chamber on a temperature controlled aluminum substrate. Film thickness (125-375 nm), ice density (0.91 ± 0.02 g/cm³) and growth rate (0.024-10 nm/s) are measured by optical interference methods using the near-normal incident light of a helium neon laser. Following deposition, the ice is annealed at 1 K/min to the point of complete sublimation. The processes of deposition, annealing and sublimation are monitored in the condensed phase using grazing-angle FTIR-Reflection Absorbance spectroscopy (FTIR-RAS) and in the gas phase using the incident water flux to a quadrupole mass spectrometer. Plots of water vapor flux versus temperature (90-130 K) give strong support for the existence of low-density amorphous ice, which manifests a flux at 128 K roughly equivalent to that of crystalline ice near 156 K [2]. With subsequent annealing, the low-density ice transforms into restrained amorphous (130-147 K), which in turn undergoes crystallization between 147-152 K. the onset of crystallization is confirmed by characteristic shifts in the FTIR-RAS spectrum [3]. Plots of desorption flux versus temperature for crystalline ice are compared to the plots of Bryson et al. [4] between 154-162 K.

Subsequent re-cooling and annealing of the crystalline ice film between 152-162 K results in further lowering of the desorption flux, substantially below the values of Bryson et al. [4]. The reason for this drop is under investigation, but may be caused by clathrate-hydrate formation. Since our films are deposited in a high vacuum chamber (base pressure $\sim 10^{-7}$ Torr) gases (roughly 3% relative to water, including $\text{CO}_2 > \text{CO} > \text{N}_2 > \text{O}_2 > \text{CH}_4$) are trapped in the ice during deposition. A small fraction of the trapped gas load is released during the phase change between amorphous and crystalline ice; the remaining (and presumably most

stabilized) gases are released as the ice sublimates, indicative of clathrate-hydrate formation [7]. Implications of these findings to astrophysical ices will be discussed.

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Satorre: IR spectra of N₂ Rich Ice Mixtures

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ABSTRACT:

The surfaces of some objects in the Outer Solar System, namely Pluto, Triton, and possibly a number of small trans-Neptunian objects seems to be dominated by frozen nitrogen mixed with small amount of methane, carbon mono and dioxide, and water. Many other, even much more complex molecules, may be as well present although not yet identified.

Because the profiles of IR bands (shape and peak position) of a given species may dramatically depend on the mixtures it is embedded in, we have studied transmission IR spectra (1.2-27 μm) of ice mixtures containing nitrogen as dominant species. Some of the studied mixtures have been also irradiated with energetic ions and their spectra collected at different irradiation doses. After irradiation samples have been warmed up and spectra collected at different temperatures. Irradiation produces molecules not originally present in the mixture (e.g., CO, CO₂ and R-OCN are produced after irradiation of mixtures N₂:H₂O:CH₄) and leaves over a complex refractory residue whose color, because of a progressive carbonization, is darker and darker as the irradiation dose increases. We have also evaluated the amount of some of the new produced species and their molecular number ratio (CO/CO₂; CO/R-OCN) as a function of irradiation dose and temperature.

The experimental results could have some interesting astrophysical implications. As an example CO and CO₂ have been detected on Triton's surface. Pluto has some CO, but less than Triton has, and no CO₂. We are investigating the possibility that CO and/or CO₂ are produced in situ by ion irradiation of appropriate frozen N₂ rich mixtures. The energetic ions trapped in Neptune's magnetic field might impact the surface and form CO₂ from CO. Pluto's surface is impacted by cosmic rays and the solar wind, but it is not subjected to bombardment by ions circulating in a nearby planetary magnetosphere, as the surface of Triton is.

It is particularly interesting the formation of CN-bearing species that once formed on primitive Solar System bodies (or incorporated in them from preexisting interstellar dust) and subsequently delivered to the Early Earth, may have contributed to the origin of life. The delivery of CN-bearing species seems to have been necessary because molecules containing the cyanogen bond are difficult to be produced in an environment that is not strongly reducing as that of the Early Earth probably was.

Selby: Luminescence Decays in UV-Irradiated Ice – The Effect of Isotopic Substitution and Temperature

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ABSTRACT:

Light emitted from the surface of a planetary body may [1, 2] be used to determine the presence and composition of surface ice. Previous work [3] has shown that when polycrystalline water ice is excited by 260 nm UV light, it emits luminescence around 420 nm. The present work extends such earlier work by examining the effect of isotopic substitution of heavy water for light water on the luminescence. In this work, the ice is exposed to UV light which is suddenly shuttered. The decay of the emitted light is then measured, decay curves being recorded at a number of temperatures, for both isotopic forms of ice. Various kinetic models are then fitted to these decay curves in order to determine the nature of the rate limiting step in the luminescence process. Recent work [3, 4] has suggested that the rate limiting step is a spin forbidden electronic transition but it is possible that it may actually be a chemical precursor step which leads to excitation. If this is the case then the precursor step may be either activation or diffusion limited, resulting in different time dependences for the decay kinetics. The time dependence equations fitted to the decay data in this work include the usual first and second order dependences but extend to first and second order fractal expressions as well as to a tunneling expression. Various combinations of these models are also being trialled.

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Schou: Sputtering of frozen CO by hydrogen ions

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ABSTRACT:

Carbon monoxide is an important constituent of the icy surfaces of the planetary bodies and of the comet comae. Even though solid carbon monoxide may only exist in the outer part of the solar system, it can be embedded in the less volatile water ice. We have studied sputtering of frozen CO by hydrogen ions for energies below 10 keV. Films of frozen CO were been deposited onto a quartz crystal microbalance at liquid helium temperature in a cryogenic setup. The mass loss and the ion current during bombardment by protons, H_2^+ and H_3^+ ions were measured. The sputtering yield turned out to depend critically on the energy and the stopping power for the ions. The yield for a 9 keV H^+ incident on solid CO was about 30 CO/ H^+ .

Sputtering of CO ice by hydrogen ion is an efficient process, and the yield is much higher than that of water ice. The reason is primarily the low sublimation energy of the solid (88 eV/CO) compared with that of water ice (450 eV/ H_2O). Our results show that the yield for CO ice increases with the square of the stopping power, similar to the behavior of the sputtering yield for water ice. It means that the particle ejection requires species from two ionization/excitation events. A complicating feature is the formation of a residue, preferentially CO_2 ice, during bombardment of solid CO. The sputtering yield depends slightly on the initial residue similar to the case of electron sputtering.

Spencer: Condensed O₂ on Europa: Discovery and Implications

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ABSTRACT:

Reanalysis of teleopic visible-wavelength spectra of the Galilean satellites taken between 1993 and 1997 reveals a weak (0.2% deep) 5770 Å absorption on Europa due to condensed molecular oxygen. The same band is seen, with about ten times the band depth, at low latitudes on Ganymede's trailing side (Spencer et al. 1995, *J. Geophys. Res.* 100, 19049; Calvin and Spencer 1997, *Icarus* 130, 505), but on Europa the band appears to have similar depth at all longitudes. A possible explanation for the distribution of O₂ on Ganymede and Europa might be that O₂ is produced everywhere by UV photolysis, or bombardment by high-energy, isotropic, magnetospheric ions, but is destroyed or released from the surface by bombardment by low-energy ions, which are deflected from Ganymede's trailing side by its magnetic field. Reconnection of field lines may allow more low-energy ions to bombard Ganymede's leading hemisphere than its trailing hemisphere, and destroy O₂ there (K. Khurana personal communication), perhaps explaining the leading/trailing asymmetry on Ganymede. The lack of strong longitudinal asymmetries on Europa, and models of bombardment of Europa would then place constraints on the ion energy range required for creation and destruction of O₂.

Trifunac: Hydrogen Atoms in Radiolysis of Water-Silica Interfaces*

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ABSTRACT:

Hydrogen atoms have an important and multifaceted role in many technologically important systems. They serve as passivators and defect reducers in materials. The remarkable radiation resistance of fused silica derives from the presence and reaction of H atoms.

Hydrogen atoms are very mobile and reactive products of ionization of water or other H bonds. The trapping and reactions of hydrogen atoms occur at very low temperatures and provide insights into the structure, trapping and reactions at interfaces.

Pulsed Time-Resolved and CW-EPR studies of trapped and reactive H atoms in ice, water-saturated zeolites and mesoporous solids, will be illustrated.

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Woon: Ab Initio Modeling Of Chemical Processes In Astrophysical Ices

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ABSTRACT:

Laboratory studies at NASA-Ames Research Center and elsewhere have demonstrated that a variety of organic species can be formed in ices under conditions analogous to those experienced in icy grain mantles that coat dust in the interstellar medium. Treatment with high energy radiation (UV, x-rays), energetic particles, or by simple heating produce different families of products in similar ices.

An overview will be presented of previous and ongoing quantum chemical calculations that complement the experimental investigations. Examples from three types of systems will be provided:

- (1) the surface reaction $H+CO$;
- (2) ice-bound reactions between formaldehyde and HCN/HNC driven by simple heating;
and
- (3) an overview of the behavior of UV-irradiated ices containing water, methanol, ammonia, and other constituents.

Yakshinskiy: Electron- and photon-stimulated desorption of K and Na from water ice films.*

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ABSTRACT:

We present laboratory studies of the electron- and photon- stimulated desorption (ESD and PSD) of atomic Na and K from thin water ice films grown on a metal substrate. These films simulate the surface of Europa. Alkali atoms are deposited onto amorphous ice (grown at 100K) or crystalline ice (grown at 150K). The measurement scheme for ESD and PSD of alkalis includes an efficient pulsed low-energy electron source, as well as mechanically-chopped UV light from a mercury arc source, a highly sensitive detector based on surface ionization, and the use of a time-of-flight technique. The energetic appearance threshold for ESD of potassium atoms from ice is $\sim 4\text{eV}$, followed by a more intense ESD desorption threshold around 25eV (O_2s excitation). The low energy threshold is more pronounced for the case of crystalline ice compared to amorphous ice, possibly due to the different ionicity of the alkali fractional monolayer as determined by X-ray photoelectron spectroscopy. We also see PSD of K for $E > 4\text{eV}$ photons. The velocity distributions for K and Na measured at 100K are peaked at ~ 500 and ~ 800 m/s, respectively, with the trailing edge extended up to ~ 3000 m/s.

The mechanism of desorption is identified as an electronically excited charge transfer from ice to alkali ion, followed by desorption; this is similar to the process proposed for ESD and PSD of the alkali / silica system [1,2]. We conclude that along with magnetospheric energetic ion sputtering processes, UV solar photons and electron fluxes with $E > 4\text{eV}$ are able to cause Na and K desorption from Europa's surface.

- 1) BVY & TEM. Nature, 400, 642 (1999).
- 2) T. E. Madey, B. V. Yakshinskiy, V. N. Ageev, R. E. Johnson. J. Geophys. Res., 103, 5873 (1998).

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