

1 INTRODUCTION

2 The lunar exosphere is generated by a variety of processes: photo-desorption from solar UV
3 radiation (PSD), solar wind ion sputtering, meteoritic bombardment, radioactive decay, and
4 thermal desorption. While remote or orbital temporal measurements provide *in situ* clues to
5 source mechanisms, individual ejection processes are more easily and deeply investigated in
6 laboratory experiments on returned Apollo samples and analogs, allowing quantitative
7 comparisons at lunar-like pressures and temperature. The importance of laboratory experiments
8 cannot be over-emphasized, providing measurements of ejection probabilities relevant to
9 exospheric formation, as well as metrics such as surface charge, surface composition and phase,
10 and meteoritic-impact plume characterization. These parameters can be convolved to describe
11 telescopic observations as well as phenomena observed at the lunar surface by orbital/lander
12 measurements, providing ground truth for models of spatial and temporal variations in the
13 exosphere. The following discussion of laboratory investigations, pertinent to the generation of
14 the lunar atmosphere, is a starting point for those interested in laboratory simulations and is by
15 no means an exhaustive review.

16 PHOTON STIMULATED DESORPTION (PSD)

17 PSD of adsorbed species occurs when solar UV photons initiate substrate-mediated excitations of
18 an electron in the adsorbed atom or in the valence band of the solid. For alkali metals such as Na
19 or K, typically adsorbed in an ionic state, the incident photon excites an electron in the mineral
20 substrate into the conduction band, which is subsequently transferred to the adsorbed Na⁺ (K⁺)
21 (Hellsing et al. 1997). Since the resulting neutral Na (K) has a larger atomic radius (2.8 Å) than the
22 ion (1.4 Å), the neutral is repelled and ejected from the surface. This process requires an alkali
23 adsorbate coverage < 0.35 monolayer (1 ML ~3 Å) to retain its ionic bond, which becomes
24 neutralized or intercalated at greater surface coverages (Yakshinskiy & Madey, 1999).

25 The mechanism for PSD of adsorbed water is similar. Water is transparent in the visible and near
26 UV (~180-900 nm), thus a substrate-mediated excitation is required to desorb H₂O. Incident UV
27 radiation produces a photoelectron in the underlying mineral, which is captured to form H₂O⁻ or
28 its dissociation product OH⁻. The excited negative ion is attracted to the mineral surface, where it
29 neutralizes. Like Na and K, the H₂O (or OH) molecule is repelled by its increased proximity to the
30 mineral substrate and ejected. PSD occurs when the surface coverage for H₂O is < 1 ML, since a
31 substrate-adsorbate interaction is required.

32 SODIUM AND POTASSIUM PHOTODESORPTION EXPERIMENTS

33 The discovery of alkali atoms in the lunar exosphere (Potter and Morgan, 1988; Tyler et al. 1988)
34 and the expectation that PSD must be a major source of Na and K precipitated a series of
35 experiments measuring the photo yield of Na. A threshold energy of ~4 eV was found for both UV
36 stimulated desorption and electron stimulated desorption (ESD) from SiO₂ is induced by an
37 electronic excitation and is measured using filtered light from a chopped Hg arc lamp (PSD; E_{max}
38 ~5 eV) and a pulsed low-energy electron gun (ESD; 0 - 200 eV) (Yakshinskiy and Madey 1999).
39 Desorbed neutral Na atoms were ionized and a time-of-flight spectrometer was used to count the
40 yield as a function of photon/electron energy. Yields were converted into a neutral Na desorption
41 cross section from SiO₂ $\sigma_d = 3 \pm 1 \times 10^{-20} \text{ cm}^2$ for ~ 5 eV photons, providing an estimated PSD flux

1 $\Phi_{\text{PSD}} = 4 \times 10^6 \text{ Na cm}^{-2} \text{ s}^{-1}$ from the lunar surface. For ESD the neutral Na desorption cross section
2 was $\sigma_d \sim 1 \times 10^{-20} \text{ cm}^2$ at 5 eV. Ionized Na^+ was observed by ESD for energies $> 25 \text{ eV}$ ($\lambda < 50 \text{ nm}$).
3 Subsequent experiments performed by Yakshinskiy and Madey (2004) using Na deposited on a
4 cooled (100 K) lunar basalt thin section yielded results similar to that for an SiO_2 substrate: a PSD
5 cross section $\sigma_d \sim 1 \times 10^{-20} \text{ cm}^2$ and suprathermal ($\sim 900 \text{ K}$) Na velocities ($v_{\text{Avg}} \sim 1 \text{ km s}^{-1}$).

6 *H_2O PHOTODESORPTION EXPERIMENTS*

7 Investigations concerning desorption of water from the surface of metals, metal-oxides, and other
8 materials have a long history and have been reviewed by a number of authors (e.g., Thiel and
9 Madey, 1987). PSD cross sections for multi-layer amorphous solid water (ASW) ice were measured
10 using Lyman- α at low-temperatures ($< 140 \text{ K}$) by Westley et al. (1995) with a highly sensitive
11 quartz-crystal microbalance (QCM) system. A QCM uses variation in the natural quartz oscillation
12 frequency to measure small mass change. These experiments found the desorption efficiency was
13 both fluence and temperature dependent. For $T < \sim 60 \text{ K}$ a yield of $\sim 3 \times 10^{-3} \text{ H}_2\text{O}$ per photon was
14 derived, corresponding to a cross section of $\sigma_d = 8 \times 10^{-18} \text{ H}_2\text{O cm}^2$ after fluences $> 2 \times 10^{18} \text{ H}_2\text{O}$
15 $\text{cm}^{-2} \text{ s}^{-1}$. Similar results were obtained for energetic electrons and protons. Yields for H_2 and O_2 ,
16 observed by residual gas analysis (RGA), were found to be small (< 10%) relative to H_2O (Westley
17 1995). Subsequent thermal desorption of the irradiated ices identified small amounts of H_2O_2 and
18 HO_2 ; OH was observed but could not be quantified. In a complementary set of experiments using
19 IR reflectance and mass spectroscopy, Öberg et al. (2009) measured a temperature-dependent
20 PSD yield $\sim 10^{-3} (1.3 + 0.032 \times T)$ molecules per photon for ASW with thickness > 8 monolayers
21 using $10^{13} \text{ photons cm}^{-2} \text{ s}^{-1}$ at fluences $1-9 \times 10^{17} \text{ photons cm}^{-2}$. The measured fraction of desorbing
22 $\text{OH}:\text{H}_2\text{O}$ was 1.4, 0.9, and 0.6 respectively, for 20, 30, and 100 K; the relative PSD fraction for H_2
23 and O_2 could not be determined with an RGA. H_2O_2 emission was not observed. For ASW ice films
24 $\sim 1.3 - 3 \text{ ML}$, the yield was found to vary with film thickness.

25 With observations of ppm concentrations of water (or hydroxyl) across the majority of the lunar
26 surface, several measurements of the photo-desorption cross section for thin layers of *adsorbed*
27 water on lunar analog surfaces have been made (e.g. Mitchell et al. 2013; DeSimone and Orlando,
28 2013a). Mitchell et al. (2013) used an excimer laser at 193 nm to photodesorb a $1.6 \times 10^{15} \text{ H}_2\text{O cm}^{-2}$
29 ($\sim 1 \text{ ML}$) ice film from an amorphous carbon substrate held at 120 K by a temperature-controlled
30 LHe cryostat. A QCM was used in conjunction with a UV spectrophotometer to measure H_2O
31 column density. A fluence-independent PSD cross section was determined, $\sigma_d = 7.4 \pm 0.5 \times 10^{-19}$
32 cm^2 , and used to calculate the H_2O desorption rate, $6.84 \times 10^{-5} \text{ s}^{-1}$, and lifetime for H_2O on the
33 lunar surface: 5 – 40 hours (Mitchell et al. 2013).

34 DeSimone and Orlando (2014a & b) measured a similar cross section for water desorption from
35 0.1 Langmuir H_2O -exposed lunar breccia. An effective water loss cross section of $\sigma_d(\text{H}_2\text{O}) \sim 7.1 \pm$
36 $1.9 \times 10^{-19} \text{ cm}^2$ was obtained and a corollary experiment found the PSD cross section for $\text{O}(\text{P}^3)$ from
37 H_2O : $\sigma_d(\text{O}) \sim 4.9 \times 10^{-20} \text{ cm}^2$. In these experiments, a lunar section held at 102 K on a LN_2 -cooled
38 finger was exposed to water vapor, providing a $\sim 0.1 \text{ ML}$ adsorbed film. H_2O molecules were
39 desorbed using a UV laser (157 nm) and probed by resonance-enhanced multiphoton ionization
40 (2+1 REMPI), a highly-sensitive, state-selective type of time-of-flight mass spectrometry. The H_2O
41 PSD cross section decreased as a function of gas exposure (film thickness) reaching $\sigma_d \sim 7.2 \pm 1.7 \times$
42 10^{-20} cm^2 at 10 ML. DeSimone et al. (2013) suggest that PSD of H_2O likely occurs by a number of

1 mechanisms, particularly recombination of surface hydroxyl photo-fragments as $\text{OH} + \text{OH} \rightarrow \text{H}_2\text{O} + \text{O}$. The ejection rate by solar photons from ice residing on the surface of Goldschmidt carter was
2 calculated as $1.6 \times 10^8 \text{ H}_2\text{O cm}^{-2} \text{ s}^{-1}$ and $7.4 \times 10^6 \text{ O cm}^{-2} \text{ s}^{-1}$ (DeSimone and Orlando, 2014a; 2014b).

4 SOLAR WIND ION IRRADIATION

5 Since the Moon has no appreciable planetary magnetic field, ions in the solar-wind (SW) impact
6 the surface at $\sim 1 \text{ keV/amu}$ during the Moon's orbit around the Earth, except during passage
7 through the Earth's magnetotail when the solar wind ion flux ($\sim 3 \times 10^8 \text{ ions cm}^{-2} \text{ s}^{-1}$) is dramatically
8 reduced. The effect of the solar wind ions on the Moon are many and include: sputtering,
9 backscattering, implantation, chemical synthesis, and enhanced diffusion. Atoms and molecules
10 sputtered or scattered from the lunar surface are a significant source of the lunar exosphere,
11 especially at high altitudes (Wurz et al. 2007).

12 *SPUTTERING YIELD EXPERIMENTS*

13 Incident SW ions transfer energy to the lunar surface causing sputtering primarily by 1) electronic
14 excitations and 2) knock-on collisions which transfer momentum to atoms in the regolith and
15 precipitate ejection of near-surface atoms or molecules. Typically, sputtered particles are neutral
16 and have energy distributions with maxima $E \leq 10 \text{ eV}$, with only a small fraction leaving as ions.
17 Below we describe experiments by 4keV He, $\sim 1\text{keV}$ H incident on lunar or analog materials;
18 penetration depths are $\sim 300 \text{ \AA}$ and $\sim 150 \text{ \AA}$, respectively.

19 Quantitative sputtering yield measurements for lunar material have not yet been acquired,
20 although total yields for mineral oxides Al_2O_3 and SiO_2 by low-energy, light ions at normal incidence
21 were measured by Roth et al. (1979) by target mass loss with a microbalance. Yields were
22 determined as $Y_S = \Delta N_T / N_I$ where ΔN_T is the change in number of target atoms and N_I is the number
23 of incident ions, measured using a Faraday Cup. For Al_2O_3 and SiO_2 , respectively, the sputtering
24 yields were $Y_S = 2.03 \times 10^{-2}$ (1 keV H^+) & $Y_S = 1.58 \times 10^{-1}$ (4 keV ${}^4\text{He}^+$) and $Y_S = 2.3 \times 10^{-2}$ (2 keV H^+)
25 & $Y_S = 9.44 \times 10^{-2}$ (4 keV ${}^4\text{He}^+$). An energy dependent universal function for the sputtering yield of
26 light ions at energies $< 20 \text{ keV}$ was fit to the data (Roth et al. 1979).

27 The effect of multiply-charged ions on the sputtering yield was measured by Meyer et al. (2011).
28 They investigated the neutral yield by incident H^+ Ar^+ , Ar^{6+} , and Ar^{9+} at 375 eV/amu from lunar
29 simulant JSC-1A AGGL using a quadrupole mass analyzer. The ejected atom mass spectra, as a
30 function of incident ion charge state and type, were compared to examine the role of potential
31 sputtering on sputtering yield. When highly charged ions approach, surface electron(s) can
32 neutralize the incoming ion by filling its outer shell(s); the excited former ion (neutral)
33 subsequently de-excites by electron emission, becoming ionized once again. Multiple fast cycles
34 of this process occur with ion approach, impact, and penetration into the surface layer, enhancing
35 the total sputtering yield (Aumayr & Winter 2004). Meyer et al. (2011) measured a fluence-
36 independent 50% yield increase for Ar^{6+} and 100% for Ar^{9+} over Ar^+ .

37 For water ice films of $\geq 100 \text{ ML}$, the sputtering yield for H_2O has been measured over a range of
38 temperatures, incident ion types, energies (sub keV to MeV), and angles of incidence. In early work
39 by Brown and colleagues (e.g., Brown et al. 1980) the ice thickness was monitored by stopping
40 power whereas later experiments use a QCM (e.g., Famá et al. 2008) with a Faraday cup for beam
41 current measurement. The total yield was found to vary as a function of ion incidence angle and

1 temperature. Famá et al. (2008) summarized 30 years of total sputtering yield measurements to
2 give a general formula for H₂O, based on the nuclear (dominant below 4keV) and electronic
3 stopping power, significant for energies \geq 4 keV He and \geq 10 keV Ar.

4 Cross sections for sputtering by 4keV He⁺ of *adsorbed* Na and H₂O on lunar soils and analogs has
5 been measured for concentrations of Na < 1 ML adsorbed on silicates: $\sigma_s(\text{Na}) \sim 1 \times 10^{-15} \text{ cm}^2$ (Dukes
6 et al. 2011) using X-ray photoelectron spectroscopy (XPS) and for *adsorbed* H₂O on lunar fines
7 using a QCM: $\sigma_s(\text{H}_2\text{O}) = 1.6 \pm 0.04 \times 10^{-16} \text{ cm}^2$ and multilayers of ice $\sigma_s(\text{H}_2\text{O}) = 3 \pm 1 \times 10^{-15} \text{ cm}^2$
8 (Mitchell et al. 2013). We note that solar-ion irradiation can also initiate chemical change and the
9 production of new species (e.g., Schaible & Baragiola, 2014), which can be subsequently ejected
10 into the lunar exosphere.

11 *ION-INDUCED SECONDARY-ION MASS SPECTROSCOPY (SIMS) EXPERIMENTS*

12 Ions sputtered from the lunar surface into the exosphere can be roughly correlated to surface
13 composition in a manner similar to secondary ion mass spectrometry (SIMS) (Johnson & Baragiola,
14 1991). Laboratory SIMS measurements by Elphic et al. (1991; 1992) with incident H⁺ (1.5 keV), He⁺
15 (4 keV), Ne⁺ (5 keV), and Ar⁺ (5 keV) on lunar simulants, using a quadrupole mass spectrometer,
16 found positive-ion sputtering yields proportional to the incident ion nuclear stopping power and
17 estimated lunar ion fluxes of $10 - 10^4 \text{ ions cm}^{-2} \text{ s}^{-1}$. Ion yields for protons were surprisingly large –
18 an order of magnitude greater than predicted by the stopping power – and roughly similar to
19 helium ion emission, while 0.1 – 1 % of the Ar⁺ yields. Positive ions with low ionization energies
20 (Na⁺, K⁺, Ca⁺, Mg⁺) are enhanced in the mass spectrum, while species with high electro-negativities
21 (O⁺, P⁺) are not observed (Elphic et al. 1991).

22 Dukes & Baragiola (2015) investigated correlations between ion yields and surface composition
23 using 4keV He⁺ incident on lunar soils with XPS and SIMS. Spectral dependence of the ejecta
24 composition on fluence equilibrated after $4 \times 10^{17} \text{ He}^+ \text{ cm}^{-2}$, as the most volatile species were
25 removed from the surface. Both atomic and molecular (NaO⁺, MgO⁺, and SiO⁺) species were
26 observed in proportion to soil stoichiometry, with relative yields dependent on secondary-ion
27 energy. No positive ion species with mass < 20 amu or > 63 amu was observed. Oxygen sputters
28 as a neutral or negative ion, observed in the negative SIMS spectrum. Ion energy distributions
29 were similar for all lunar soils, rising rapidly with energy to a maxima at \sim 5 eV for Na⁺ and the
30 molecular ions, \sim 7.5 eV for Fe⁺, and \sim 10 eV for Mg⁺, Al⁺, Si⁺, Ca⁽⁺⁾, Ti⁺, before decreasing slowly
31 (Dukes & Baragiola, 2015).

32 THERMAL DESORPTION

33 Thermal desorption from the lunar surface occurs due to solar heating, and the exosphere
34 generally follows a \sim 29.5 day diurnal pattern for gases that condense on the lunar surface at low
35 temperatures. Temperature cycling between 25K (in permanently shadowed regions or nighttime)
36 to 400 K affects desorption of most species, excluding H₂, He, Ne, Ar, CO, O₂, and N₂ which do not
37 condense. Experiments on thermal desorption of volatiles trapped in lunar materials were begun
38 in the Apollo era by direct heating of returned lunar samples (e.g., Gibson & Johnson 1971), with
39 recent work on measurement of activation energy for adsorbed water and alkali metals on lunar
40 analogs and basalt by thermal desorption (Poston et al. 2015; Yakshinskiy et al. 2000).

1 *THERMAL DESORPTION OF ADSORBED SODIUM EXPERIMENTS*

2 For adsorbed alkali atoms, such as Na and K at surface coverage < 1 ML, thermal desorption (<
3 1000 K) is facilitated by electron transfer from the substrate, similar to the mechanism for PSD
4 discussed earlier. Therefore, the majority desorb as neutrals, although a small fraction of ions are
5 detected at higher temperatures. Temperature programmed desorption measurements (TPD),
6 utilizing a mass spectrometer, by Yakshinskiy et al. (2000) for Na on 100 Å SiO₂ atop Re, show that
7 the onset for thermal desorption of Na for fractional monolayers is ~500 K, with the maximum
8 rate at ~700 K. For Na concentrations > 1 ML, thermal desorption occurs lower temperatures with
9 the maximum rate at ~330 K and a broader, less intense peak at ~700 K for Na adsorbed to SiO₂.

10 *THERMAL DESORPTION OF ADSORBED H₂O EXPERIMENTS*

11 The thermal stability of adsorbed H₂O and OH on lunar analogs was studied by Hibbitts et al. (2011)
12 and on Apollo soils by Poston et al. (2015). Both measured thermal desorption of ~10 ML of
13 adsorbed and chemisorbed water from powder (or soil) samples using TPD and quadrupole mass
14 spectrometry at < 5 x 10⁻⁹ Torr. After outgassing to 750K the powder was cooled to ~110 K (165 K
15 for Apollo soils), water vapor was deposited and then heated at a constant rate. The onset of
16 sublimation is observed at ~150 K from ice layers atop the H₂O /mineral interface, then a broad
17 peak due to chemisorbed H₂O (maxima at 225 K); for Apollo soils the peak of the desorption rate
18 for chemisorbed H₂O was ~250 K with a rapidly decreasing tail. An activation energy of ~1.5eV for
19 highland soil was determined (Poston et al. 2015).

20 Sack & Baragiola (1993) measured the sublimation rate for crystalline (~140 – 170 K) and
21 amorphous ice films (< 130 K) by TPD using a QCM, finding an enhancement in sublimation rate
22 (~3x) for low temperature films. This exponentially decayed within hours to the crystalline rate.
23 For 140 – 170 K a temperature-dependent sublimation flux was empirically described.

24 METEORITE AND MICROMETEORITE BOMBARDMENT

25 LADEE and Kaguya measurements of sodium in the lunar exosphere suggest both short and long
26 term enhancements in column density for exospheric constituents produced by meteoritic impact
27 (Colaprete et al. 2016; Kagitani et al. 2010). In the laboratory, micrometeorite impacts are
28 simulated by 1) hyper-velocity impact in test laboratories where single projectiles (typically) are
29 accelerated to < ~10 km/s to impact a target and 2) by laser ablation using high-intensity pulsed
30 lasers (~10¹⁰ W cm⁻²). It should be noted that there are fundamental differences in energy transfer
31 mechanisms, vapor volume and temperature between the two processes.

32 *HYPERVERELOCITy IMPACT AND PULSE LASER IRRADIATION OF MINERAL EXPERIMENTS*

33 In characteristic hypervelocity impacts (~ 5 km/s), Sugita et al. (1998) used 6 mm quartz pellets
34 impacting dolomite. The impact vapor temperature was found to correlate with the velocity
35 component normal to the target surface, v_{perp} , as $T \propto (v_{\text{perp}})^{0.3}$. Spectrographs with fast CCD
36 cameras found the vapor plume consisted of three types of photoemission: blackbody radiation
37 (heat from impact), molecular band, and atomic line emission. The intensity of the line emissions
38 (Ca, Mg, Na, CaO, and MgO) to the single-temperature blackbody continuum (3000 – 4000 K) in
39 the impact plume is largest immediately after impact, following a Boltzmann distribution for vapor
40 temperatures of 4000 – 6000 K.

1 Mukhin et al. (1989) used mass spectrometry and pulsed-laser heating of mafic minerals, rocks,
2 and meteorites in He or H₂ atmospheres to determine the composition of released gases. For
3 basalt, gabbro, peridotite, and augite the release of H₂, N₂, CO, CO₂, and hydrocarbons was
4 observed and quantified. Kadono et al. (2002) investigated the temperature, radius, and
5 composition for a vapor cloud generated by pulsed 1.06μm laser irradiation of basalt in low-
6 vacuum (0.15 Torr). A vapor temperature of ~2000 K was observed 100 ns after impact and found
7 to decrease with time. From the vapor pressure and density, Kadono et al. (2002) estimated the
8 laser simulation was equivalent to an impact between two basaltic bodies of relative speed ~120
9 km/s. A number of groups have collected laser-ablation vapor plumes on various lunar-analog
10 substrates (i.e. Loeffler et al 2008, Sasaki et al. 2001), analyzing redeposition products in terms of
11 mineralogy and spectral reflectance, rather than as a source of exospheric vapor.

12 SUMMARY AND ACKNOWLEDGEMENTS

13 This chapter briefly encapsulates laboratory work done to better understand the formation and
14 evolution of the lunar exosphere. Laboratory experiments that compliment remote observations
15 and guide computer simulations are critical to understanding fundamental physical and chemical
16 mechanisms operating at the lunar surface, including interaction between lunar exosphere and
17 regolith and the generation of the lunar environment. Fundamental studies by the authors
18 concerning the effects of space weathering, measurement of lunar sputtering yields, electron
19 emission and charging of the lunar surface are underway and supported by NSF-Astronomy (C.D.)
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