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The Influence of Temperature and Photobleaching on Irradiated Sodium Chloride at Europa-like Conditions

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Abstract

Europa's leading-hemisphere chaos regions have a spectral feature at 450 nm that has been attributed to absorption by crystal defects in irradiated sodium chloride, known as F-centers. Some discrepancies exist between the laboratory data of irradiated sodium chloride and the observations, including a ∼10 nm shift in central wavelength of the F-center band and the lack of the prominent 720 nm absorption on Europa from M-centers, which result from the coalescence of pairs of F-centers. Here, we perform irradiation experiments on sodium chloride in an attempt to understand these discrepancies. We show that careful control of the temperature of the sample at 120 K yields F-centers with an absorption wavelength comparable to that of Europa. In addition, we measure the effect of photobleaching—the destruction of F-centers by photons—and show that at the energetic particle and photon flux on Europa, an equilibrium will be reached where only a modest F-center absorption develops. The density of F-centers never reaches high enough values for the creation of secondary M-centers. Our experiments predict that F-centers grow during the night on Europa in the absence of photobleaching and then partially decay during the daytime. We show observations from the Hubble Space Telescope consistent with this prediction. All observations of the 450 nm F-center on Europa are now consistent with laboratory measurements of sodium chloride, confirming the presence of this salt on Europa.

Unified Astronomy Thesaurus concepts: Galilean satellites (627); Europa (2189); Planetary surfaces (2113); Surface composition (2115)

1. Introduction

Jupiter's moon Europa has a young, icy surface that undergoes high-energy radiation bombardment and covers a salty, liquid-water ocean (Kivelson 2000; Paranicas et al. 2001, 2002). The salts in the ocean presumably derive from the interaction between the liquid water and the silicate seafloor (Kargel et al. 2000), and the composition of the salts offer insight into the manner of this interaction. While no direct access to the ocean is currently possible, Europa's geologically young chaos regions have likely been resurfaced by material ultimately derived from the interior ocean and thus could provide information on its composition (Carr et al. 1998; Glein & Shock 2010; Schmidt et al. 2011; Soderlund et al. 2014). On the trailing side of Europa, these chaos terrains are bombarded by sulfur plasma, confusing the interpretation of their initial composition; the leading-side chaos terrains are thus the most likely to retain signatures of the original composition.

Trumbo et al. (2019) recently reported Hubble Space Telescope (HST) observations that showed the existence of an absorption feature at 450 nm in Europa's leading-side chaos terrains and proposed that this feature is due to sodium chloride salts. While salts are generally indistinct in the visible wavelength range, irradiation of alkali halides, in particular, produces multiple defects and dislocations within the crystal structure, giving rise to distinct "color-center" absorptions. For NaCl, the most prominent color center is the F-center, occurring at approximately 450 nm (Mador et al. 1954).

F-centers occur when radiation ejects a chlorine atom from its location in the crystal lattice and an electron fills the hole. More complicated color centers also form, including M-center absorption at 720 nm, which appears when the density of F-centers is sufficiently high that pairs begin to coalesce. This F-center coalescence can eventually lead to macroscopic sodium colloids which broadly absorb in the 580 nm region. F-centers can be destroyed by absorption of photons within the 400–500 nm band of the absorption feature in a process called photobleaching, which excites the electron into the conduction band (Herman & Wallis 1955).

Experiments performed in order to understand the possibility of NaCl color centers on Europa (Hand & Carlson 2015; Poston et al. 2017; Hibbitts et al. 2019) suggested that irradiation at conditions similar to those on Europa would form both F-centers and M-centers. The HST data, however, did not completely match the laboratory data. In the laboratory, the central wavelength of the F-center was measured to be 460 nm, rather than the ∼450 nm seen on Europa, and the HST data showed no hint of the predicted absorption near the 720 nm M-center. Trumbo et al. (2019) noted that when the irradiation of the NaCl stopped in the Poston et al. (2017) experiments, the F-center wavelength quickly shifted blueward as the absorption rapidly decayed. They hypothesized that irradiation at the lower flux level seen on Europa could lead to the shorter wavelength feature. They further hypothesized that the lack of M-centers on Europa is the result of solar photobleaching which would destroy F-centers at nearly the same rate as their creation through irradiation. This destruction would lead to an equilibrium with a low number of F-centers that would not have a high enough density for M-centers to form.

Here, we perform experiments to test these hypotheses. Our goal is to determine if the differences in appearance between

Table 1
Experiments

	Current Density (nA cm^{-2})	Electron Flux (electrons cm^{-2} s^{-1})	450 nm Irradiance (mW m^{-2} nm^{-1})	Dose Rate (eV/16 amu yr^{-1})	Temperature (K)
Low	12	8×10^{10}	6.0	2.5×10^{3}	120
Low	12	8×10^{10}	⋯	2.5×10^{3}	120
–	48	3×10^{11}	6.0	1×10^{4}	120
Medium	505	3×10^{12}	6.0	1×10^{5}	120
Medium	505	3×10^{12}	⋯	1×10^{5}	120
Medium	505	3×10^{12}	6.0	1×10^{5}	120 (no indium foil)
Medium	505	3×10^{12}	⋯	1×10^{5}	200
High	3410	2×10^{13}	6.0	7×10^{5}	120
High	3410	2×10^{13}	⋯	7×10^{5}	120
Europa	n/a	n/a	78	10	120

Note. The energy deposited is for the top 1.2 μm micron of the sample. For the experiment without indium foil the temperature of the salt is unknown. The irradiance of our spectral lamp is estimated at 450 nm. For Europa, the photon flux and temperature are given for equatorial noon.

the HST spectra and the laboratory data can be reconciled with the interpretation that the ∼450 nm absorption feature seen on Europa is indeed due to irradiated NaCl.

2. Method

Our experiments were designed to measure the spectral changes in sodium chloride from electron and photon bombardment at temperatures similar to Europa. For each experiment, fresh hydrated sodium chloride powder, USP (CAS-7647-14-5), was pressed into a layer of indium foil inside of a copper sample cup. Excess salt was brushed off to ensure good thermal coupling between the remaining grains and the sample cup. The sample cup was then loaded onto a cold finger inside of a Kimball Physics ultra-high vacuum chamber. The chamber was pumped down to 10^{-8} torr using an Agilent ID3 backing pump and TwisTorr 84 molecular turbo pump. The cold finger was cooled with a Janis ST-400 liquid nitrogen cryostat. Once the desired temperature was reached, the grains were irradiated with 10 keV electrons from a Kimball Physics EGG301 electron gun. The current is measured with a Kimball Physics Faraday cup mounted on a linear actuator that can be inserted into the electron beam path. Electron currents used in the different experiments and their relationship to the approximate energy flux on Europa are shown in Table 1. We obtained spectra of the samples from 300 to 1000 nm by illuminating the sample via an external lens fed by a fiber optic connected to both a stabilized deuterium and a stabilized tungsten-halogen lamp from Thorlabs. We illuminated the sample at 45° from the surface normal and collected the diffuse reflection 90° from the specular direction and 45° from the surface normal. Since NaCl is nearly featureless through the spectral range, unirradiated salt at the desired temperature was used as a spectral reference for the our samples.

For our series of experiments the electron flux and illumination conditions were varied. In our initial experiments, we simply tested the system to ensure that we could create and measure F- and M-centers. Our initial electron flux, as measured in the Faraday cup, was 2×10^{13} electrons cm^{-2} s^{-1}. Since the penetration depth of 10 KeV electrons is around 1.2 μm (Hand & Carlson 2015) such a flux gives the equivalent of 7×10^{5} eV (16 amu)$^{-1}$ deposited each year into the NaCl. The energy deposited on the apex of Europa's leading hemisphere is estimated to be 10 eV (16 amu yr)$^{-1}$ (Nordheim et al. 2018). Our initial experiments are at a dosage rate approximately 70,000 times that of Europa. The true dosage rate on Europa is both poorly determined and variable, so this ratio is only an approximation. Additional experiments were run at fluxes 7 times lower and nearly 300 times lower than our initial experiment. These three settings are referred to as high, medium, and low flux. It should be noted that even the low flux is ∼250 higher than that of Europa. Figure 1 shows the results of our first experiment at high flux. F-centers are quickly created and become saturated within approximately 30 minutes. As the density of F-centers increases, coalescence of these defects begins to form M-centers, which begin to absorb at 720 nm by 30 minutes. By the end of the experiment, both F- and M-center absorptions are prominent.

In some experiments, we test the effects of photobleaching by leaving our spectral lamp illuminated during the irradiation. While we have no means of measuring the absolute flux density of our spectral lamp inside of the chamber at 450 nm, we estimate this parameter by using the spectral curve and total output provided by the manufacturer as well as the illumination spot size on the sample. We find the irradiance of the lamp at 450 nm to be 6 mW m^{-2} nm^{-1}. This irradiance is approximately 8% of the flux density at Europa from solar illumination.

3. Experiments

The visible spectrum of the leading-side chaos regions on Europa differs from laboratory spectra of irradiated NaCl in two important ways. First, the Europa feature appears at ∼450 nm, rather than the 460 nm wavelength found for NaCl F-centers in the laboratory (Hand & Carlson 2015; Poston et al. 2017). Second, the laboratory spectra all show the formation of an M-center at ∼720 nm, which the HST data strongly rule out. We perform a set of experiments to explore these phenomena.

3.1. Wavelength Shift

Trumbo et al. (2019) noted that, in experiments by Poston et al. (2017), the F-center spectral feature quickly decayed and shifted blueward when irradiation ceased. They hypothesized that the higher irradiation flux for the laboratory spectra caused the mismatch in wavelength between the Europa and the laboratory spectra. Our first set of experiments was designed to test that hypothesis.

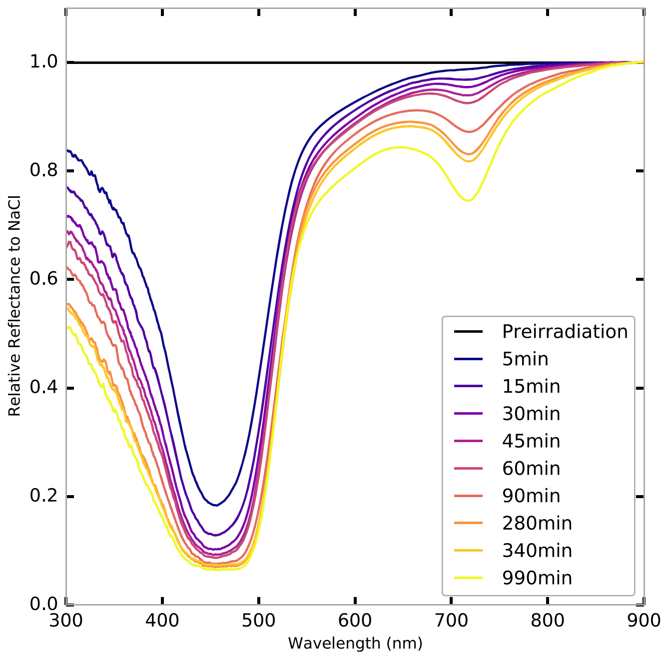

Figure 1. Spectra of irradiated sodium chloride at 120 K with our high irradiation dosage and no photobleaching. M-center growth is observed after F-center saturation.

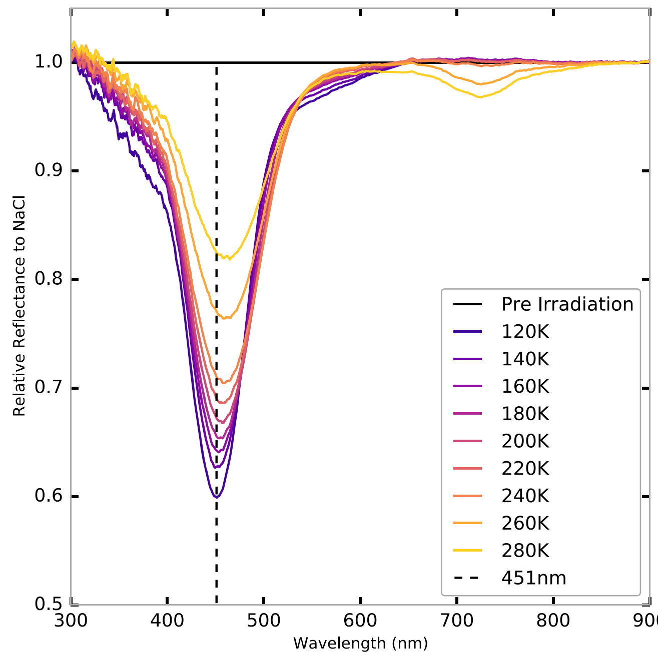

Figure 2. Irradiation at 120 K with systematic warming of the sample. This shows the redward shift of the F-center with temperature. The central wavelengths from 120 to 280 K are 451.2, 453.1, 454.1, 456.1, 457.6, 459.0, 460.5, 461.9, and 463.4 nm. The minimum values were found by doing a parabolic fit to the F-center range (400–500 nm) and then finding the wavelength minimum of the parabola.

We first attempted to reproduce some of the key results of Poston et al. (2017). We irradiated a NaCl sample at a temperature of 120 K at our medium flux level, which is approximately 6 times that of the initial Poston et al. (2017) irradiation, with no photobleaching. This flux is approximately 10^4 times the flux level at Europa. We irradiated for 2 hr, periodically obtaining spectra, to measure the growth of both the F-center and the M-centers. This 2 hr irradiation is equivalent to 2.3 yr on Europa. We observed a central wavelength for the F-center of 451 nm, which is significantly shifted from the Poston et al. (2017) results and much closer to the spectra from Europa (Figure 2), even though our flux level was 6 times higher. At the end of the irradiation sequence, the samples were left in the dark at 120 K overnight. In contrast to Poston et al. (2017), we observed no change in the spectrum after ceasing irradiation. We then systematically warmed the sample, allowing the sample to stabilize at each new temperature, and obtained a spectrum at each, in order to examine the effects of temperature. As noted by Schwartz et al. (2008), the wavelength of the F-center systematically shifts redward with increasing temperature. As well as shifting, the depth of the F-center decreases with increasing temperature. By 240 K the peak of the F-center absorption is at 460 nm, and at 260 K, M-centers begin to spontaneously grow, as the higher temperatures allows the F-centers to mobilize and begin to coalesce (Figure 2).

The difference between these experiments and the results of Poston et al. (2017) are striking. We obtain a significantly shorter wavelength for the F-center, and we see no spontaneous spectral change postirradiation until we purposely raise the temperature. We hypothesize that these differences are due to a lack of adequate thermal coupling between the salt grains and the cold finger in the Poston et al. (2017) experiments. The angular grains have little contact with each other or with the cold finger and are exposed to thermal emission from the walls of the experimental chamber. These grains could be at a significantly higher temperature than the copper sample cup and the cold finger. By embedding the samples in indium foil, as was done in the present experiments, the thermal coupling is greatly increased. To test this theory, we irradiated salts in the same manner as before, but without pressing the sample into indium foil. We found that the central wavelength shifted to 460 nm, and the growth of both the F-center and M-center occurred on much faster timescales when compared to spectra at 120 K (Figure 3). As a final test, we irradiated indium-pressed thermally coupled salt grains at 200K. For these spectra, the central wavelength was observed at 458 nm. There was no discernible shift in the central wavelength postirradiation but a slight growth in the M-center was observed postirradiation. The peak of the absorption for these 200 K experiments was blueward of both the Poston et al. (2017) spectra as well as our experiments without pressing the salt into the indium foil (Figure 3). Based on these experiments, we hypothesize that the grains in the Poston et al. (2017) experiments were not well coupled to the cold finger. We believe they were approximately ∼240 K, based on the central wavelength and the growth of the M-center. This higher temperature would naturally lead to the redward shift of the F-center and the postirradiation spectral changes observed in those experiments but not observed in our thermally coupled 120 K experiments. We conclude that the shift in the F-center between Europa and laboratory data is not due to the increased irradiation in the laboratory, but rather from the inadvertently elevated sample temperature in the previous laboratory data. For thermally coupled samples at 120 K, the central wavelength of 451 nm is within the uncertainty of the positional measurement of the feature on Europa. The position we observed also coincides with the central wavelength previously measured for these temperatures (Mador et al. 1954; Schwartz et al. 2008).

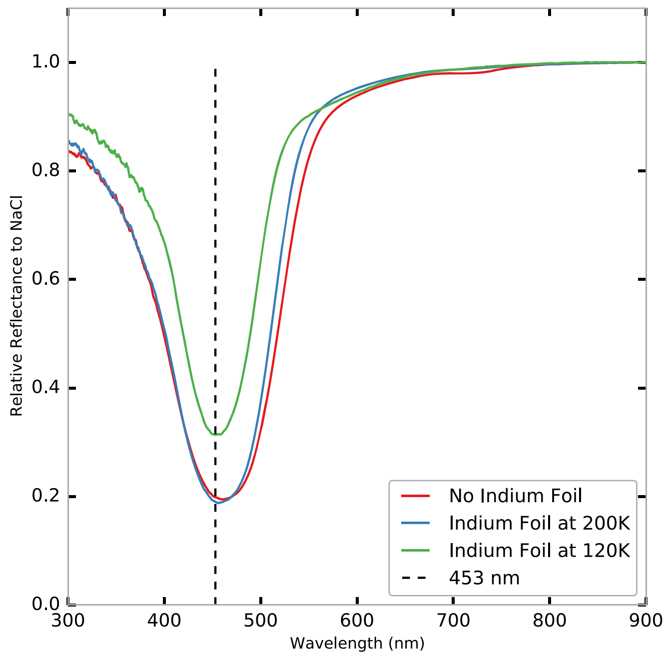

Figure 3. Spectra of NaCl irradiated at medium irradiation for 15 minutes. Significant differences are seen in samples with indium foil at 120, and 200 K, and without indium foil. F-center growth is significantly faster for the 200 K and no indium foil samples, while the F-center wavelength is also shifted redward. For the no indium foil sample, M-center growth can already be seen. We conclude that the sample sitting on a 120 K cold finger but without indium foil to thermally couple it is even warmer than 200 K.

3.2. M-Center Formation

The second critical difference between the laboratory and HST data is the strong growth in the laboratory of the M-center absorption feature located at 720 nm compared to its nondetection from HST. Trumbo et al. (2019) hypothesized that photobleaching, the destruction of F-center color centers by photons, could suppress the formation of F-centers sufficiently that M-centers, which require a certain density of F-centers in order to coalesce, might never form. To test the magnitude of this photobleaching effect, we performed two sets of experiments. The first was a series of experiments with and without photobleaching at three different irradiating levels. For photobleaching we use the same lamp as is used for spectral reflection. Our lamp provides roughly 8% of the photon flux density in the 400 to 500 nm wavelength span of the F-center compared to noon at Europa's equator. At all irradiation levels, photobleaching slowed the growth of the F-centers, as can be seen in the comparison between photobleached and nonphotobleached spectra with otherwise identical conditions in Figure 4. To additionally examine this effect, we measured the mean absorption depth of the F-center from 425 to 475 nm as a function of time for each of our irradiation experiments. As seen in Figure 5, the average absorption depth of the F-center is dependent on both electron flux and whether or not there is photobleaching. The growth rate of the F-centers, without photobleaching, is approximately linear until saturation occurs and the formation of M-centers begins. This linear region is poorly resolved in the medium and high irradiation experiments, but if we take the 5.6% absorption growth in the first 20 minutes of the low irradiation experiment to be typical of the quasilinear growth regime, we find a growth rate of 17% hr^{-1}. Scaling by the irradiation flux gives consistent results for the

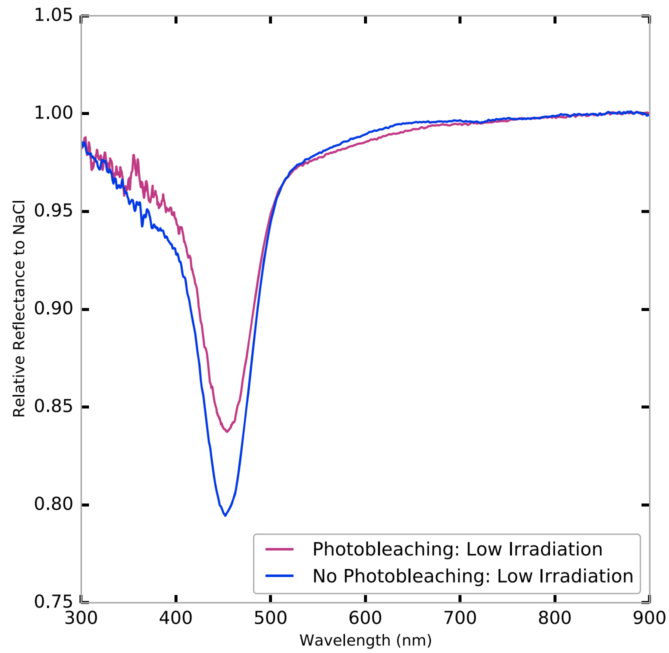

Figure 4. Spectrum of NaCl irradiated at low irradiation for 40 minutes with and without photobleaching. The central wavelength is the same for both spectra.

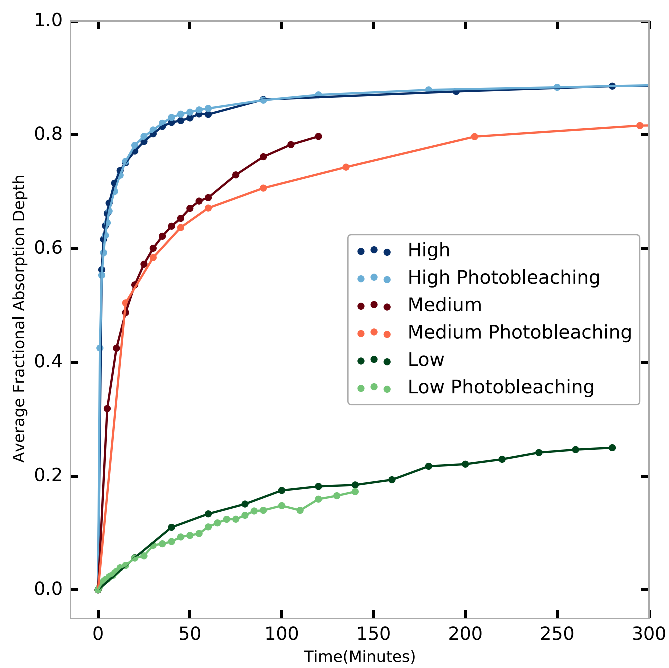

Figure 5. The average fractional absorption depth of the F-center from 425–475 nm vs. time of irradiation for our three irradiation levels, with and without photobleaching. In all cases, photobleaching slowed the growth of the F-centers, though the lamp is too weak to completely balance even the lowest electron irradiation level.

medium and high irradiation levels. We thus assume that for small F-center absorption depths, the growth of the absorption depth, A, can be described as a simple linear growth, $dA/dt = aF$. Here, F is the irradiation flux in units of eV $(16\ \mathrm{amu\ yr})^{-1}$, where the flux at Europa is ~10, and a is a constant, here found to be 6.8×10^{-5} in the same units as F and with t in hours. At the flux of Europa, a ~7% absorption

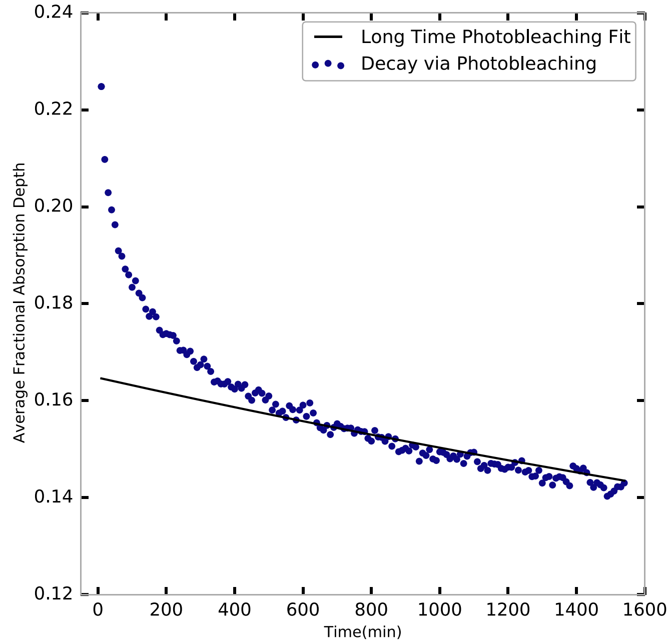

Figure 6. The average fractional absorption depth of the F-center from 425 to 475 nm with photobleaching postirradiation.

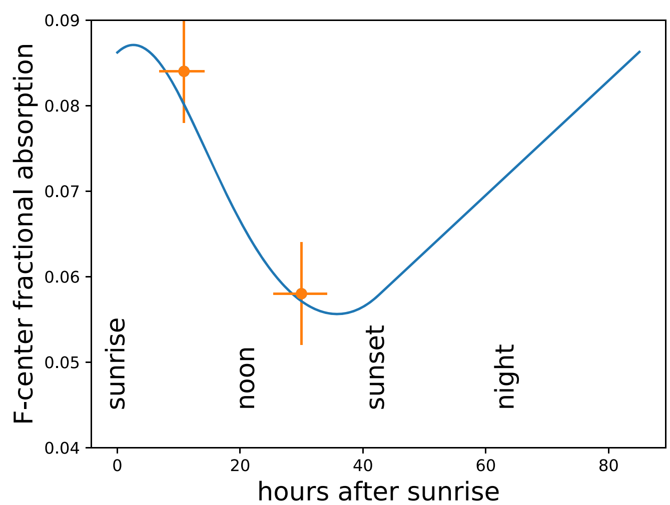

Figure 7. A simple numerical model showing the F-center fractional absorption as a function of Europa's 85 hr rotation period, including the effects of continuous irradiation on Europa with solar flux dependent photobleaching during the daytime. The F-center grows over the course of the night and begins decaying soon after sunrise before recovering after sunset. The data points show the absorption depths of two HST measurements of identical regions within Tara Regio taken at different local times. The horizontal error bars show the geographic extent of the observed region. The F-center decay predicted by the model is observed on Europa.

feature would grow in a little more than one orbital period in the absence of competing effects.

It is difficult to simultaneously match the photon and electron flux at Europa with our experimental setup. Even our lowest level of irradiation is ∼250 times that experienced at Europa, while our photobleaching photon flux density is ∼13 times *less*. Moreover, every time we take a spectrum, we are photobleaching for the ∼1 minute duration of the spectral exposures. Thus, even the unbleached spectra have actually had some amount of photobleaching. For these reasons, it is difficult to extrapolate the results of Figure 5 to estimate the effect at Europa.

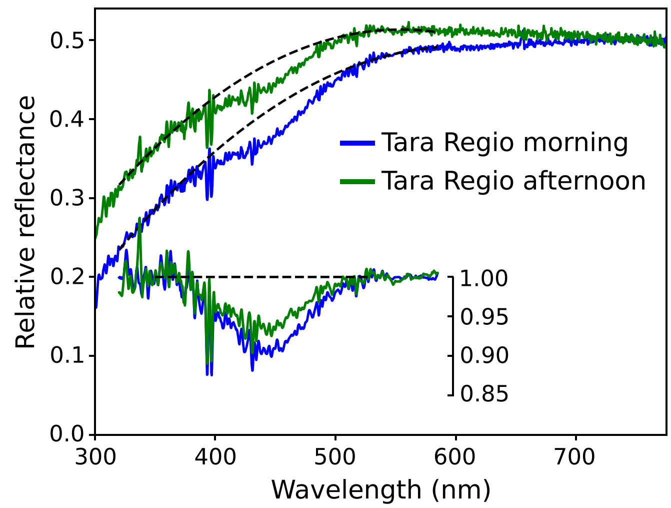

Figure 8. HST observations of overlapping regions of Tara Regio in the morning and in the afternoon. Dashed lines are second-order polynomial continuum fits, which facilitate band-strength comparison. The continuum-removed absorption bands are shown below. The observed decay in the F-center absorption is consistent with the photobleaching decay predicted here.

To attempt to obtain a better estimate of the effect of photobleaching on Europa, we performed one additional experiment. We irradiated a sample at four times our low irradiation flux for 10 minutes, leading to an F-center with a fractional absorption depth of about 20%, approximately three times that seen on Europa. We then ceased irradiation and began photobleaching. The F-center absorption decayed rapidly for the first few hours (Figure 6). By the time it reached values more comparable to those seen on Europa, the decay slowed, and we fit this section of the curve to the theoretically derived and experimentally verified equation for photobleaching of lightly irradiated NaCl, $dA/dt = -bIA^2$, where A is the fractional absorption depth of the F-center, I is the intensity of the light, and b is a constant. (Herman & Wallis 1955). We find a value of 0.035 for the product bI for our lamp, which scales to 0.46 for the noon time equatorial solar flux at Europa.

We used this photobleaching equation and an assumed linear growth rate of F-centers with irradiation to estimate an equilibrium value of F-center absorption depth. We model the F-center growth rate as $dA/dt = aF - bIA^2$. In steady-state and assuming continuous photobleaching at the average equatorial day/night irradiance, this equation predicts an absorption depth of 6.8% for Europa. This calculated value is remarkably similar to the depth seen in the HST data. Crucially, such an equilibrium value for the F-center is much lower than that seen to be required for formation of M-centers. In reality, photobleaching is not constant on Europa, but ranges from zero during the night to a maximum at noon. Incorporating this changing photobleaching rate into a simple numerical model which accounts for nighttime and sinusoidally varying illumination in the daytime suggests that, in equilibrium, the F-center absorption depth would grow to 8.7% overnight, decay to 5.6% by the end of the day, and regrow overnight (Figure 7).

Intriguingly, the HST data on Europa show evidence for this precise photobleaching effect. While Trumbo et al. (2019) report only average values for the F-center strength across the disk, we note that a portion of Tara Regio was observed both in the morning and in the afternoon. Reproducing their data reduction, but examining overlapping spectra separately, we find that the morning absorption depth is indeed deeper than that of the afternoon. (Figure 8). These absorption depths are well fit by our simple time-dependent mode. Given the crudeness of the modeling and the difficulty of extrapolating laboratory conditions to those at Europa, we believe that the extremely close agreement between the precise values of the model and of the data must be mainly coincidental. Nonetheless, we find the fact that the relative change in the F-center absorption depth from HST agrees with the model to be an encouraging sign that our extrapolations to Europa's surface conditions and crude modeling are capturing the real behavior at Europa.

4. Conclusions

We used new laboratory data with carefully controlled temperature and photon illumination conditions to examine the discrepancy between the spectrum of the Tara Regio region on Europa measured with HST and the spectrum of irradiated NaCl found in previous laboratory work. We found that in our experiments irradiated NaCl grains, which are thermally coupled to a 120 K cold finger by pressing them into indium foil, develop F-center absorptions at 451 nm, matching the spectral feature on Europa, rather than at the 460 nm wavelength seen in previous experiments. Absorption at 460 nm is, however, seen when samples are placed on the cold finger without pressing into the foil and when thermally coupled samples are heated to 240 K. We conclude that previous experiments had poor thermal coupling between the cold finger and the angular salt grains resting on a cold finger, resulting in a raised sample temperature of approximately 240 K. At the appropriate temperature, the band position of laboratory-irradiated NaCl at 451 nm falls within the uncertainty of the band position on Europa.

Previous laboratory data also predicted the presence of M-center absorption at 720 nm on Europa for typical irradiation doses expected, but the HST data put strong upper limits on the existence of such an absorption. Here we found that photobleaching—the destruction of F-centers by photons—can nearly precisely balance the irradiation-induced creation of F-centers, leading to a low equilibrium value for F-center density and a weak absorption strength, similar to that seen on Europa. This low equilibrium density is well below the density required for pairs of F-centers to begin to coalesce to form M-centers, explaining the lack of this additional absorption on Europa.

Our model for F-center creation and destruction predicts that the absorption will grow overnight in the absence of photobleaching and partially decay during the day under solar illumination. We show that morning and afternoon spectra of Tara Regio, when examined separately, show this effect at a similar level as predicted.

These new laboratory experiments show that the visible spectrum of Tara Regio is consistent with the presence of irradiated NaCl, including the wavelength of the F-center feature, the lack of an M-center absorption at 720 nm, and the change in the strength of the F-center feature over the course of the day. The existence of NaCl in a chaos region on the leading hemisphere of Europa strongly suggests that sodium and chlorine are dominant components of the subsurface of Europa.

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