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# Scattering of atoms and molecules off a magnesium oxide surface

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**Abstract**

Neutral particle imaging in the low energy range will, when employed in future spacecraft missions, potentially contribute to enlarging our knowledge about the origin and evolution of our universe. The main difficulty of a neutral particle detector in this energy range is that the incoming neutrals must first be ionized such that they can be mass- and energy-analyzed by conventional methods. In this paper we report on the first observation of the formation of negatively charged ions upon reflection from a magnesium oxide surface. Experiments were performed at two different setups, one of them including a time-of-flight measurement of the specularly reflected particles, the other one allows angle resolved detection. We measured high fractions (12–30%) of negative ions and less than 1% of positive ions when scattering $O^+$, $O^0$, $O_2^+$ and $O_2^0$ at 90–3000 eV primary energy per atom off a MgO surface. When scattering $H^+$, $H_2^+$ and $H_2^0$ at 300–1500 eV per atom off the MgO surface, 3–7% of the particles were converted into negative ions and 2–10% into positive ions and when scattering $C^+$and $C^0$ at 1500–2000 eV per atom around 3% of the particles were converted into negative ions and 3% into positive ions. Our results strongly suggest a complete memory loss of the incident charge state. Scattered molecules dissociated always completely. The mean energy loss was approximately proportional to the incident particle energy except for $O^-$ ions where an increased loss at lower energy was observed. MgO therefore meets the most important requirements for application on a space platform. © 2002 Published by Elsevier Science B.V.

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## 1. Introduction

Knowledge about the origin and evolution of our solar system, our galaxy and the universe can be gained by determining the composition of matter. The measured abundances of the elements and isotopic ratios help constrain cosmological models. The composition of our solar system reveals interstellar matter of the protosolar nebula some 4.5 billion years ago. A sample of the present-day galaxy with reliable observations of a number of important elemental and isotopic abundance ratios is missing. Unlike the ionized component of the interstellar gas, which is excluded from penetrating inside the heliosphere by the solar wind plasma,

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the neutral component of the interstellar gas (mainly H, He and O are expected) can penetrate deeply into the heliosphere and therefore they might be measured even as close to the Sun as the Earth orbit. This measurement yields direct information about the composition of the local interstellar medium. Moreover, energetic neutral particles (expected energy range is 0.1–5 keV), which are produced by charge-exchange between interstellar neutral gas and heliospheric particles can be measured. A two-dimensional image of the neutral fluxes helps infer the interaction processes between the local interstellar medium and the heliosphere. We expect to detect mainly atomic particles (mainly H, He, O) since they are much more abundant than molecules in the local interstellar gas or in planetary magnetospheres. However, in a neutral particle detector the incoming neutrals must first be ionized such that they can be mass-analyzed by conventional methods. Neutral particle imaging in the low energy range requires efficient ionization methods since the expected fluxes are very small. Surface ionization was identified as the only viable ionization technique to meet the requirements concerning ionization efficiency for the energy range of 10 eV to 1 keV within the limitations imposed by the resources (space, weight, power, etc.) available on a satellite [1]. It was concluded that the detection efficiency should exceed 1%, should be uniform over large areas, and should exhibit good longterm stability during the mission duration. Surface ionization introduces new demands on the design of the mass spectrometer and requires the development of new analyzer elements with matched ion optical properties. An instrument meeting these demands has been described recently [2,3] and is currently operating successfully on the IMAGE satellite [4]. Surface ionization was studied extensively for low-work function surfaces [5–7] with the low-work function of the surfaces established by coating a typical metal with an alkali or an alkaline-earth over-layer [8,9]. Preparing such coated metal surfaces with sufficient quality and reproducibility in an instrument on a spacecraft turns out to be quite involved [10]. Thus, we focused our research on insulating surfaces [11–15], which yield reasonable results without surface preparation in space. The insulating surfaces are much easier to handle than the rather involved coated tungsten surfaces, which is of great importance for an application on a spacecraft. A recent review on neutral particle detection techniques and their application in space instrumentation can be found in [16]. In this work we present the experimental results for the negative and positive ion fractions we obtained when scattering hydrogen, oxygen and carbon particles off a MgO surface. In addition, we report on the energy loss the projectiles experience upon scattering.

## 2. Experimental setup

The measurements reported here were made in part at the University of Bern and in part at the University of Osnabrück. The two setups will both be described briefly. More detailed information on the experimental setups at the University of Bern and University of Osnabrück can be found in [11–13] and [17], respectively. The investigated scattering surface was a bulk MgO(1 0 0) single crystal polished to an RMS roughness of less than 5 nm (1 nm typical), as measured by optical means. The orientation accuracy of the (1 0 0) plane relative to the polished surface was better than 0.5°. Charge fractions were all measured at the University of Osnabrück except for the measurements using primary $O_2^+$ particles which were also done at Bern using the ILENA apparatus.

The experiment ILENA at the University of Bern consists of an ion source, a beam guiding system, a sample stage with housing and a detection unit. All these units are contained in a single vacuum chamber pumped by a turbomolecular pump. Molecular ions of oxygen were used because they can be produced far more efficiently in this system. The impact angle of the ion beam on the surface under investigation can be chosen between 90° and 0° with respect to the surface normal. The reflected beam is recorded with a two-dimensional position-sensitive MCP detector with a viewing angle of ±12.5° in both azimuthal and polar direction. A retarding potential analyzer (RPA) consisting of three grids is mounted in front of the MCP detector. The detector unit, including

the RPA, is shielded electrostatically and can be rotated independently from the converter surface around the same axis. The outer grids of the RPA are grounded to shield the inner grid, which can be biased to suppress positive ions. An additional grid in front of the MCP detector at negative potential with respect to the MCP detector serves to reject secondary electrons originating from the preceding grids and the converter surface. The MCP detector may be floated to a high negative voltage with respect to the converter surface to eliminate negative particles. After baking out the vacuum chamber a residual gas pressure of $4 \times 10^{-8}$ mbar is achieved. During operation the pressure may rise into the low $10^{-7}$ mbar range as a result of the test gas leaking into the ion source chamber. The sample can be heated in order to remove adsorbates from the surface.

The experimental system JUSO [11,17] at the University of Osnabrück is an UHV system with a base pressure in the low $10^{-9}$ mbar range. Primary ions are extracted from an ion plasma source and mass analyzed by a 90° sector field magnet. With an electric deflection system in front of the analyzing magnet the primary beam can be pulsed to allow time-of-flight (TOF) measurements. The scattering surface is mounted onto a three axis manipulator equipped with electron beam heating. A thermocouple is used for temperature control. Downstream from the scattering surface, the TOF tube is mounted under a fixed scattering angle of 10°. Scattered ions are separated from scattered neutral particles by a postacceleration voltage. In the detector a microsphere plate (MSP) is used. The aperture angle for detection is 1.2°. Neutral primary particles are produced by neutralization of the primary ion beam in a gas cell that is placed after the analyzing magnet and before the scattering chamber.

## 3. Results

In the scattering experiments we used hydrogen, oxygen and carbon as primary particles. We produced beams of neutral and ionized atomic as well as molecular particles. We analyzed the composition of the reflected beams, the angular and energy scatter of the reflected beam, as well as the average energy loss after reflection from the surface.

*Hydrogen*: We used primary $H_2^0$, $H_2^+$ and $H^+$ particles in the energy range 300–1500 eV per atom. Fig. 1 shows a TOF spectrum for a 935 eV per atom primary $H_2^0$ beam reflected from the MgO surface at a scattering angle of 10° and postaccelerated with ±1700 V (JUSO measurement). The charged particles therefore appear at smaller TOFs, corresponding to their higher kinetic energies. In the scattered particle flux we can identify $H^0$, $H^+$ and $H^-$ particles. We never detected any charged hydrogen molecules in the scattered beam in the TOF spectrum nor did we observe any recoil atoms. Moreover, we did not observe a $H_2^0$ peak, which would appear at the same TOF as the $H^0$

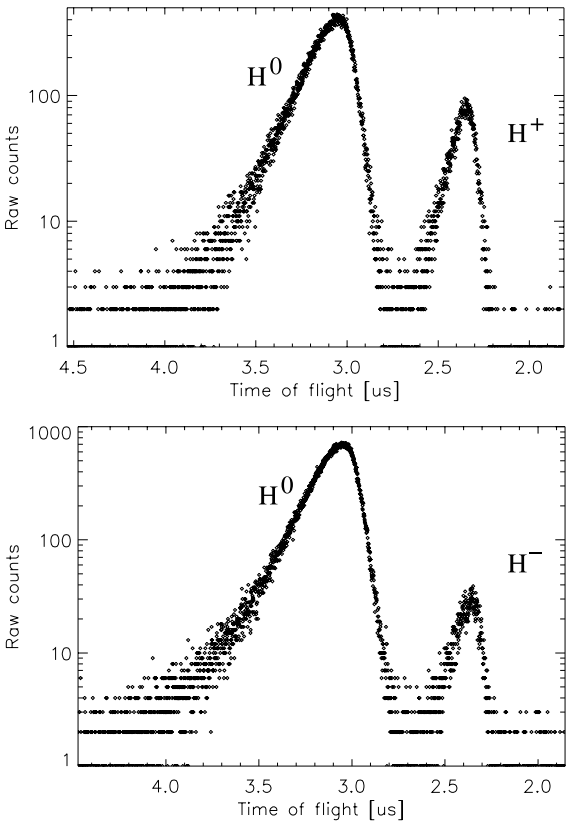


Fig. 1. TOF spectrum for primary $H_2^0$ molecules at 935 eV energy per atom and 1700 V postacceleration voltage. Upper panel: $H^+$ fraction, lower panel: $H^-$ fraction. No surviving $H_2^0$ molecules were observed.

peak, but would be narrower and thus clearly distinguishable [18,19]. The peak for each species in the TOF spectra was then transformed into an energy spectrum. The data were corrected for the relative detection efficiency of the MSP detector in the JUSO apparatus [20,21]. From the energy spectra we extracted the yields of neutral particles and ions, the energy loss and the energy straggling towards lower energies by fitting a theoretical peak shape to these data [21,22]. The used fit-function is basically a convolution of a gaussian peak with an exponential function. Fig. 2 depicts the fits to the measured energy distributions of the outgoing particles for a 1468 eV primary $H^+$ beam reflected from MgO at a scattering angle of 10°. After reflection the energy distributions were essentially the same for neutral, positively and negatively charged hydrogen particles. This corresponds well to the results of the mean energy loss, which came out the same for all primary hydrogen particles.

*Oxygen*: Primary $O_2^0$, $O_2^+$, $O^0$ and $O^+$ particles from ~400 to ~3000 eV per atom were scattered off the MgO sample. Fig. 3 shows a TOF spectrum for a 1443 eV per atom primary $O_2^+$ beam scattered off the MgO surface and accelerated with +2000 V. Only the $O^0$ and $O^-$ peaks could be observed (Fig. 3). By comparing Figs. 1 and 3 one can already see

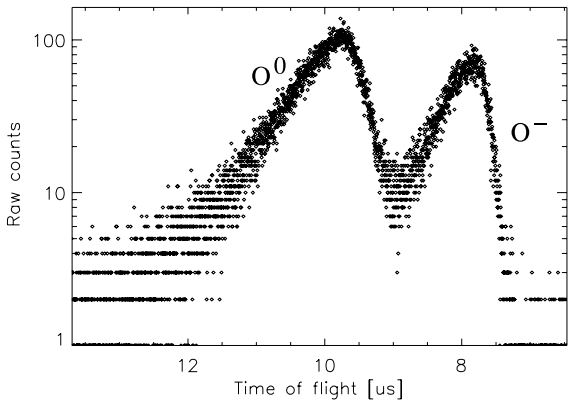


Fig. 3. TOF spectrum for primary $O_2^+$ ions at 1443 eV per atom energy and a +2000 V postacceleration voltage.

that the $O^-$ yields are substantially larger than $H^-$ yields. For a negative postacceleration voltage a few $O^+$ ions could be detected. We never found any molecular peak in the TOF spectra, which means that all oxygen molecules dissociated during the scattering event on the MgO surface. For oxygen similar corrections for detection efficiency as for hydrogen were applied. Measurements for oxygen show no difference in the energy distributions for outgoing neutral and negatively charged particles. This can be seen from Fig. 4, which depicts

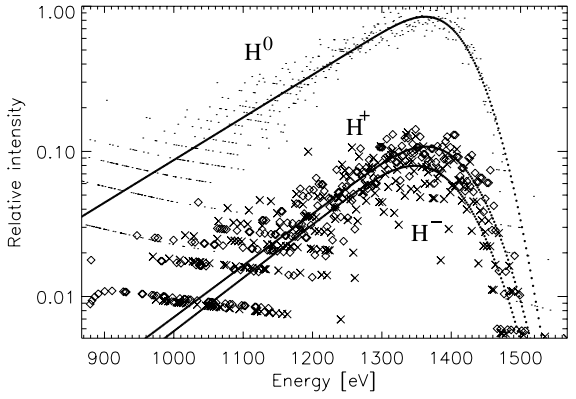


Fig. 2. Fits to the measured energy distributions for neutral (top line), positive (middle line) and negative (bottom line) hydrogen particles after reflection for a $H^+$ primary beam at 1468 eV. The slight difference in energy loss upon scattering between positive and negative outgoing ions can be seen clearly. Within the experimental uncertainty the shapes of all three curves are the same.

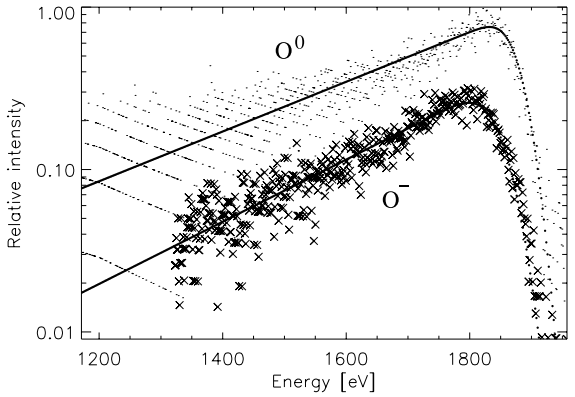


Fig. 4. Fits to the measured energy distributions for neutral (upper line) and negative (lower line) oxygen particles after reflection. The primary beam was a 1965 eV $O^+$ beam. The difference in energy loss upon scattering between neutral and negative outgoing ions can be seen clearly but there is no significant difference the the shape of the curves.

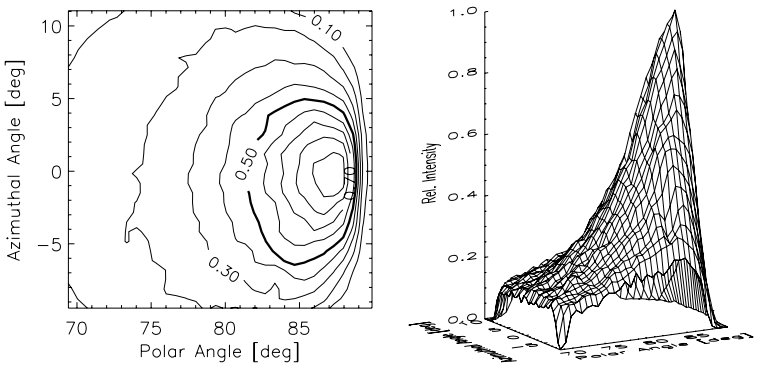


Fig. 5. Angular scattering for a 390 eV per atom $O_2^+$ primary beam at 85° incidence angle (as measured from the surface normal) measured at the University of Bern. The measured scattering at FWHM is 11° and 8° in azimuthal and polar direction, respectively. The estimated reflection efficiency is about 20%.

the fits to the measured energy distributions of the outgoing particles for an incoming 1936 eV $O^+$ beam. The shape of the energy distribution of the negative oxygen atoms correspond well to that of the neutrals. The angular scatter in azimuthal and polar direction was measured with the ILENA apparatus and is shown in Fig. 5 for a 390 eV per atom $O_2^+$ primary beam. It was 11° and 8° at FWHM in azimuthal and polar direction, respectively (see also Fig. 15 below). We also estimated the reflection efficiency, i.e. the total number of particles scattered inside the detection area of ±12.5° divided by the total number of particles incident on the sample. We obtained about 20%. In the ILENA apparatus surface charging had to be considered because of beam currents being the several orders of magnitude higher than at the JUSO setup. A filament placed close to the MgO sample provided the electrons needed to neutralize the positively charged surface. These electrons caused additional background that was subtracted before further processing of the data.

*Carbon*: We also scattered primary $C^0$ and $C^+$ particles from ~1100 to ~2000 eV per atom off the MgO sample. The obtained charge state fractions are small, about half the yields we found for hydrogen primary particles.

### 3.1. Negative and positive ion yields

Figs. 6–8 depict the summary of the measured charge state fractions, for positive and negative

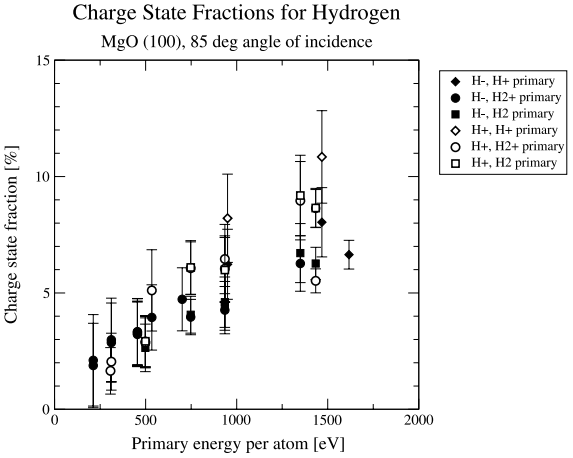


Fig. 6. Positive and negative charge state fraction for hydrogen scattered off a MgO surface as a function of primary energy per atom. The figure shows the fractions of outgoing $H^-$(filled symbols) and $H^+$ (open symbols). The charge state fractions do not depend on the primary particle type.

ions, using hydrogen, oxygen and carbon primary particles over the investigated energy range. As a general trend we find that the charge state fraction increase with the energy of the primary particle. Moreover, the nature of the primary particle beam (neutral or ionized) does not appear to have an effect on the charge state yields. For hydrogen at 300 eV only about 2% of the detected particles were positively charged. This fraction increased with energy and at 1500 eV we measured about 10% positive ions. At 300 eV about 3% of the detected particles were negatively charged. Again,

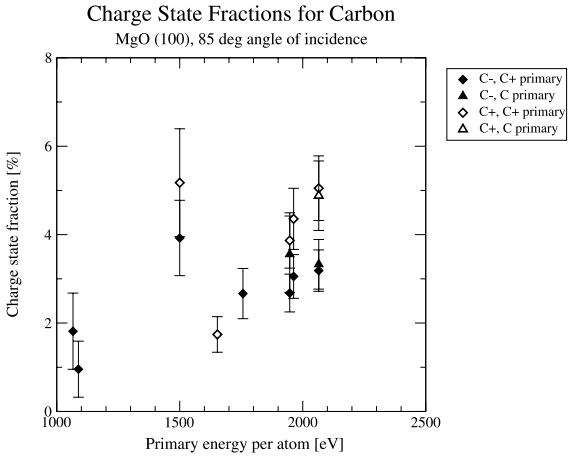


Fig. 7. Positive and negative charge state fraction for carbon. The figure shows the fractions of outgoing $C^-$ (filled symbols) and $C^+$ (open symbols). The charge state fractions do not depend on the primary particle type.

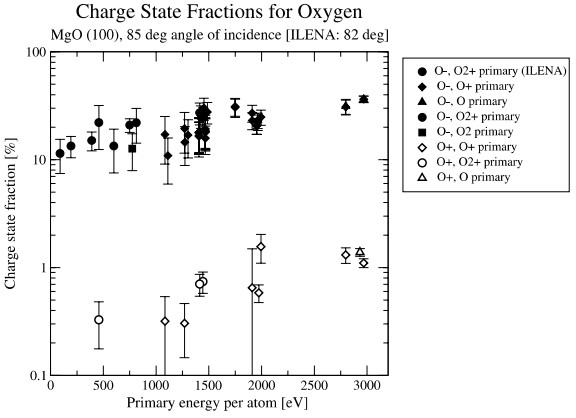


Fig. 8. Positive and negative charge state fraction for oxygen. The figure shows the fractions of outgoing $O^-$ (filled symbols) and $O^+$ (open symbols). The charge state fractions do not depend on the primary particle type.

this fraction increased with energy and at 1500 eV we measured about 7% negative ions. Within the measurement uncertainties we got the same charge fractions for $H_2^0$, $H_2^+$ and $H^+$ primary particles. For oxygen primary particles below 1500 eV per atom we find very low positive ion yields. Less than 0.5% of the detected particles were positively charged. This fraction increased with energy somewhat and at 3000 eV we measured about 1% positive ions. The negative charge state fraction at 500 eV was about 15–20%. This fraction increased with energy and at 3000 eV we measured about 30% negative ions. Within the measurement uncertainties we got the same charge fractions for $O_2^0$, $O_2^+$, $O^0$ and $O^+$ primary particles. Different orientations of the [0 0 1] direction of the surface with respect to the incoming beam while keeping the reflection angle constant did not change the charge fractions either. In the JUSO apparatus we varied the sample temperature between 20 and 150 °C using the electron beam heater. No differences in the charge state fractions for all the investigated primary particles could be observed between a cold and a hot sample. The negative charge state fraction using primary $O_2^+$ was also measured using the ILENA apparatus. These measurements were also corrected for the different detection efficiencies of the MCP detector in the ILENA apparatus for neutrals and ions [23–25]. No correction for partially dissociated molecules was necessary because the JUSO results suggested a complete dissociation of the primary molecules upon reflection of the MgO surface. At 90 eV per atom we obtained negative charge state fractions from 5% to 10%, this fraction increased up to 20% at 750 eV per atom (Fig. 8). The results obtained at the ILENA apparatus and the JUSO apparatus agree well with each other within the measurement uncertainties. We note that at the JUSO apparatus the incidence angle with respect to the surface normal was about 85° given by the fixed scattering angle of 10°. At the ILENA apparatus we varied the incidence angle from 78° to 87° and we did not observe a significant dependence of the negative charge state fraction for $O_2^+$ primary particles on the incidence angle. For carbon primary particles about 3% of the detected particles were positively and about 3% negatively charged. We got the same charge fractions for $C^0$ and $C^+$ primary particles.

### 3.2. Energy loss

The obtained values for the energy loss are shown in Figs. 9–11 for the neutral and in Figs. 12–14 for ionized scattered particles, respectively. The energy loss was in the range between 10% and 30% depending on species and energy of the

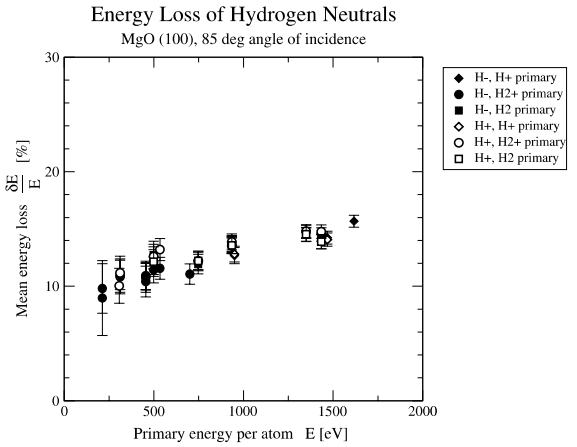


Fig. 9. $H^+$ ions (diamonds), $H_2^+$ molecular ions (circles), $H_2^0$ molecules (squares) and $H^0$ atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $H^0$.

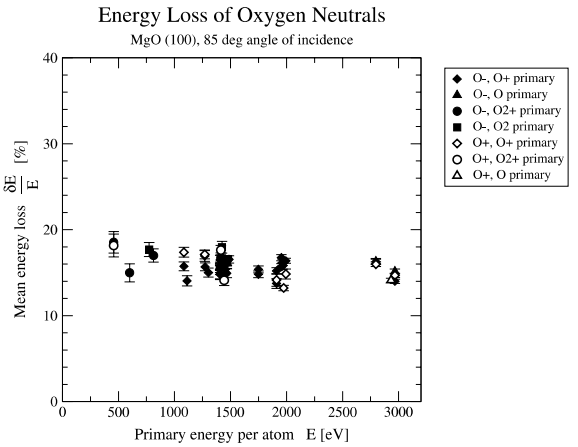


Fig. 11. Primary $O^+$ ions (diamonds), $O_2^+$ ions (circles), $O_2^0$ molecules (squares) and O° atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $O^0$.

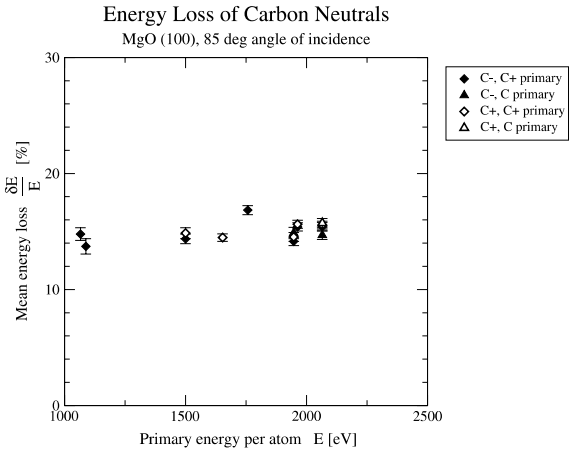


Fig. 10. $C^+$ ions (diamonds) and $C^0$ atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $C^0$.

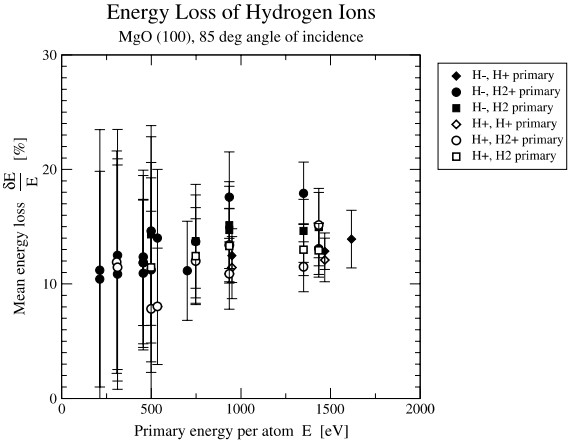


Fig. 12. Primary $H^+$ ions (diamonds), $H_2^+$ ions (circles), $H_2^0$ molecules (squares) and $H^0$ atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $H^-$ (filled symbols) and $H^+$ (open symbols). The energy loss does not depend on the primary particle type.

primary particle. For hydrogen the mean energy loss for neutrals amounts to 10% at 500 eV per atom and increases to 13% at 1500 eV per atom. For negative ions the mean energy loss seems to be slightly higher and for positive ions slightly lower than for neutrals (Figs. 9 and 12). For oxygen we found an approximately constant energy loss of 15% for neutral oxygen atoms after reflection from the surface (Fig. 11). For both negatively and positively charged oxygen atoms the mean energy loss ranges between 30% at 500 eV per atom and 15% at 3000 eV per atom (Fig. 14). For carbon we found no significant difference in the energy loss of neutral and charged atoms after reflection from the surface. The neutral atoms and the ions both

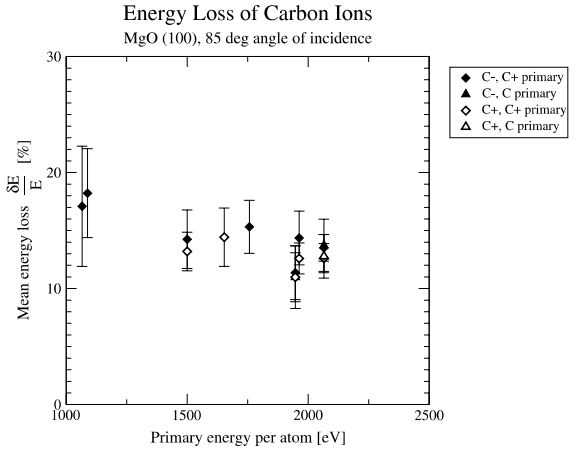


Fig. 13. Primary $C^+$ ions (diamonds) and $C^0$ atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $C^-$ (filled symbols) and $C^+$ (open symbols).

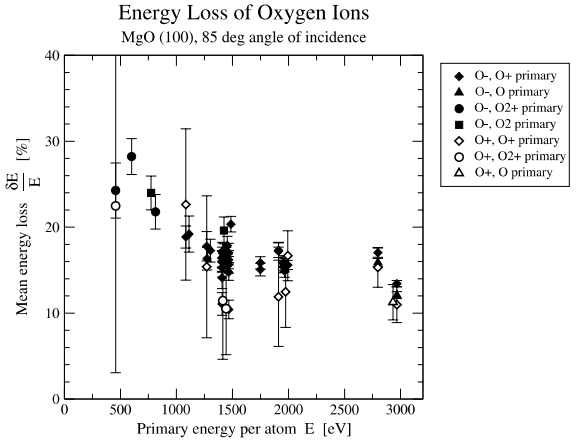


Fig. 14. Primary $O^+$ ions (diamonds), $O_2^+$ ions (circles), $O_2^0$ molecules (squares) and $O^0$ atoms (triangles) were scattered off a MgO surface at 10° scattering angle. The figure shows the mean energy loss measured for outgoing $O^-$ (filled symbols) and $O^+$ (open symbols). The energy loss does not depend on the primary particle type.

lost 15% of their primary energy at 1500 eV per atom (Figs. 10 and 13).

### 3.3. Angular scattering

A strong dependence of the angular scattering on primary particle energy and angle of incidence

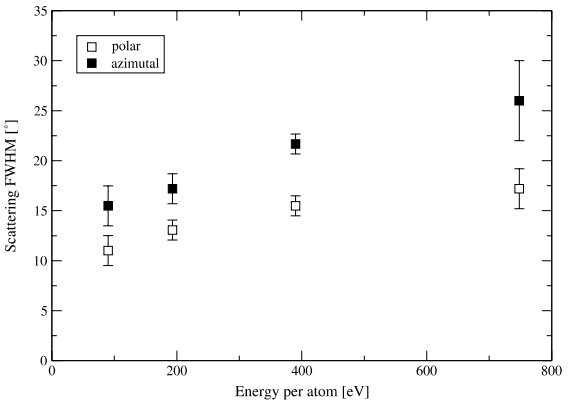


Fig. 15. Energy dependence of angular scattering obtained when scattering a $O_2^+$ primary beam of a MgO surface at an angle of incidence of 82°. The FWHM in azimuthal and polar directions are shown as defined in Fig. 5.

was found. Measurements were made using $O_2^+$ primary beam. The scattering increases considerably with energy (Fig. 15) and also with steeper angles of incidence. At 390 eV per atom and 85° angle of incidence the scattering was 11° in azimutal and 8° in polar direction, whereas an angle of incidence of 82° the scattering increased to 16° in azimutal and 11° in polar direction, respectivly (Figs. 5 and 15).

## 4. Discussion

When scattering $H_2^+$, $H_2^0$ or $H^+$ off the MgO surface at the JUSO apparatus, we found that the measured fractions of negative and positive ions in the scattered beam did not depend on the primary particle. This suggests a complete memory loss of the incident particles. A complete memory loss of the incident charge state was reported for other projectile-target combinations, in particular for scattering ions off metal surfaces [7], for scattering ions off an insulating surface [11] and also for scattering oxygen particles off a semiconductor like silicon [26]. The memory loss can be explained by effective neutralization of the ions when approaching the surface. On the incoming trajectory incident molecules could dissociate due to electronic excitation, resulting from capture of the

neutralizing electron into an antibonding state. The measured fractions of positive and negative ions in the scattered particle flux are then most probably established on the outgoing trajectory. When scattering $C^+$ and $C^0$ off the MgO surface at the JUSO apparatus, we found that the measured fractions of negative and positive ions in the scattered beam were independent from the primary particle charge state as well. This indicates a complete neutralization of the projectile before impact. The charge state yields were about half of what we obtained for hydrogen at a similar primary energy per atom. At least the negative ion fraction appears to be low considering that $C^-$ has an affinity level of 1.26 eV [27], which is almost as big as the oxygen affinity level of 1.46 eV [27]. Moreover, hydrogen has a lower affinity level of 0.754 eV [27] but has higher negative ions yields [28]. When scattering $O_2^+$, $O_2^0$, $O^+$ and $O^0$ off the MgO surface at the JUSO apparatus, we could not identify any surviving molecules in the scattered beam. We also found no significant dependence of the charge state fractions from the primary particle used. The ion yields for primary $O_2^+$ obtained at the ILENA apparatus agreed well with those obtained at the JUSO apparatus within the measurement uncertainties. The uncertainties of the JUSO results are higher than those of the Bern results because we had to consider the relative detection efficiency values at two very different energies (for the neutral particles and for the postaccelerated ions). The relative errors might amount to about 20% for energies below 1000 eV and become smaller with increasing energy because the relative detection efficiency curve flattens. We could not find a significant dependence of the charge state fractions of the background pressure. In the ILENA apparatus the pressure was about $10^{-7}$ mbar. This situation is what has to be expected onboard a satellite as the pressure inside an instrument will be of the same order. In the JUSO apparatus the measurements were performed at a residual gas pressure of about $10^{-9}$ mbar. We also considered the effects of surface charging. In the JUSO apparatus we could not find a difference in the charge state fractions between a cold 20 °C and a hot 150 °C sample, assuming that at the higher temperature the increased conductivity of MgO will reduce the surface potential. Not finding a difference might be a result of the low beam intensity of less than $3 \times 10^5$ particles $cm^{-2}\,s^{-1}$. This beam intensity is about 100 times lower than lowest usable intensity at the ILENA apparatus where charging effects were visible in the spatial distribution of the reflected particles. In the ILENA apparatus this was compensated with a higher background pressure of about $5 \times 10^{-7}$ mbar at the cost of an increased background signal and a higher uncertainty in the results. At the JUSO we were able to rotate the sample in azimuth direction. We did not observe any changes in the ionization efficiency while rotating the sample around an axis normal to the surface as mentioned in [29]. The lack of azimuthal effects is possibly due to the surface roughness of the MgO crystal which inhibits the observation of such effects at the geometry used in our experiments. The sample used had a typical surface roughness of 1 nm as specified by the supplier [30]. At JUSO we never observed any recoil atoms in the TOF spectra, which is a strong evidence that the surface was clean. The mean energy losses found for hydrogen, carbon and oxygen particles scattered off a MgO surface are mostly caused by inelastic scattering. The elastic energy losses upon binary collisions with a surface atom would not exceed about 1%. The energy losses measured are in the range between 10% and 30%, which are rather high compared to results reported in the literature for other particles reflected from metallic surfaces [17,31,32]. The fact that the mean energy loss of hydrogen and carbon particles does not depend on whether we used primary $H_2^+$, $H_2^0$, $H^+$, $C^+$ or $C^0$ is consistent with the proposed complete dissociation and complete neutralization of the molecules on the incident particle trajectory. The measured higher energy loss for negative oxygen ions compared to neutrals cannot be explained by current theories. The difference cannot be due to surface charging because the loss did not depend on the sample temperature. Moreover, surface charging would also be observable when scattering hydrogen and carbon, which is not the case. We also considered the relative detection efficiency but also this cannot account for the different energy losses.

## 5. Conclusions

We measured high fractions (about 12–30%) of negative ions and less than 1% positive ions when scattering oxygen particles at 90–3000 eV primary energy per atom off a MgO surface. When scattering carbon particles at 1100–2000 eV per atom about 3% of the particles were converted to positive and 3% to negative ions. When scattering hydrogen particles at 300–1500 eV per atom off the MgO surface, about 3–7% of the particles were converted into negative ions and about 2–10% into positive ions. There is strong evidence for a complete memory loss of the incident charge state. Scattered hydrogen molecules dissociated almost completely (>99%) and we observed no surviving oxygen molecules. The variation of the ion fractions with energy will probably be small within an environment as present in a satellite instrument. The mean energy loss was approximately proportional to the incident particle energy. We measured 12% for outgoing $H^+$, $H^0$ and $H^-$, 15% for $C^0$ and $C^+$, 15% for $O^0$and between 15% and 30% for $O^-$ for energies between 400 to 3000 eV per atom. We conclude that MgO is a promising material for application on a space platform. It meets the most important requirements and seems better than AlN [12] concerning the conversion of hydrogen atoms.

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