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Scattering of light molecules from thin Al_2O_3 films

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ABSTRACT

Molecular oxygen and hydrogen ions were scattered at grazing incidence from various thin Al_2O_3 films. The energy of incident particles was varied from 390 to 1000 eV. For scattered positive oxygen ions, negative ion fractions of up to 17% were recorded. For scattered positive hydrogen ions, the negative ion fractions reached up to 2%. These findings qualify thin films of Al_2O_3 as possible candidates for use as charge state conversion surfaces in neutral particle sensing instruments, which will work in space.

1. Introduction

The interaction of atomic and molecular particles with insulating surfaces has been researched extensively in recent years [1–10]. Reports of relatively high fractions of negative ions resulting from scattering of positive atomic and molecular ions off insulating surfaces suggested possibilities for several new applications. Among these applications we use this process for efficient detection of 10 eV–2 keV neutral particles in interplanetary and interstellar space [11–13]. The proof of concept for this detection technique has already been demonstrated in space with the IMAGE satellite mission [14]. The mass spectrograph used there is designed to detect low energy neutral atoms. It uses a conversion surface of volatile adsorbates on a highly polished, poly-crystalline tungsten substrate to convert a fraction of the incoming neutral atoms to negatively charged ions [14,15]. For more recent missions, such as IBEX from NASA, diamond-like carbon surfaces are used as conversion surfaces because of their better long-term stability and higher negative ion yield [9,10], whereas for future missions, such as BepiColombo from ESA, several different materials are still under investigation. Because of technical reasons, the conversion surfaces will be exposed to space from the very beginning of the mission. During separation of rocket stages, a small amount of Al_2O_3 will be created and, although unlikely, might be adsorbed on the spacecraft, including the conversion surfaces. Therefore, as a worst-case scenario, we had previously tested a rough film of Al_2O_3 on both a graphite substrate as well as a highly polished Al_2O_3 single crystal for ionization efficiency and scattering properties [16]. We found fractions of negative ions high enough to suggest the use of Al_2O_3 as a possible new material for conversion surfaces, but we also observed very broad scattering cones and surface charging, which is why we decided to repeat these measurements in this study with smooth thin Al_2O_3 films.

Interstellar gas is expected to consist mainly of H and He with traces of O, N, C and Ne [17], but since the objective was to analyze the suitability of Al_2O_3 films to work as conversion surfaces in space we focused our tests on oxygen and hydrogen. Molecular ions were used because they can be produced far more efficiently than atomic ions in our test system. The impact of using positively charged molecular ions on the results is discussed in detail below.

The measurements were done at moderate vacuum conditions, i.e. in the low 10^{-7} mbar range, which mirror the conditions within a typical particle sensing satellite instrument shortly after launch. From the European Space Agency's (ESA) ROSETTA mission it is known that several weeks after launch, the pressure in the vicinity of the spacecraft drops to the low 10^{-9} mbar range, and into the 10^{-10} mbar range after few months. The pressure inside space instruments with small openings to vent to the outside, such as most particle instruments, is expected to be at least an order of magnitude higher than the pressure outside the spacecraft. Because of internal outgassing, pressures in the 10^{-8} mbar range

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are expected to persist in space particle instruments more or less indefinitely.

2. Experiment

The surfaces tested are thin Al_2O_3 films on silicon substrates. Some of the substrates were etched with HF prior to the coating process. The film thicknesses vary from 172 to 25 nm.

Measurements were made with the ILENA apparatus at the University of Bern, Switzerland. The energy range of incident particles chosen for these measurements was 390–1000 eV. For all measurements the angle of incidence was 82° with respect to the surface normal. The scattering angle was determined by rotating the detector to maximize the scattered peak. The experimental setup will be described briefly. More detail on the experimental setup can be found in [11] and [18]. Fig. 1 displays a sketch of ILENA. It consists of an ion source, a beam filter and beam guiding system, a sample stage with housing and a detection unit. All these units are contained in a single vacuum chamber pumped by an ion pump. For the measurements reported here an impact angle of 8° with respect to the surface plane has been chosen. The reflected beam is recorded using a two-dimensional position-sensitive MCP detector with a viewing angle of ±12.5° in both azimuthal and polar directions, as demonstrated in Fig. 2. 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. A grid at the entrance of the MCP detector at negative potential with respect to the first MCP 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 in order to vary the transmission threshold for negative particles. After baking out the vacuum chamber at 80 °C overnight, a residual gas pressure of 5×10^{-8} mbar is achieved. During operation the pressure may rise into the low 10^{-7} mbar range as a result of test gas leaking into the ion source chamber.

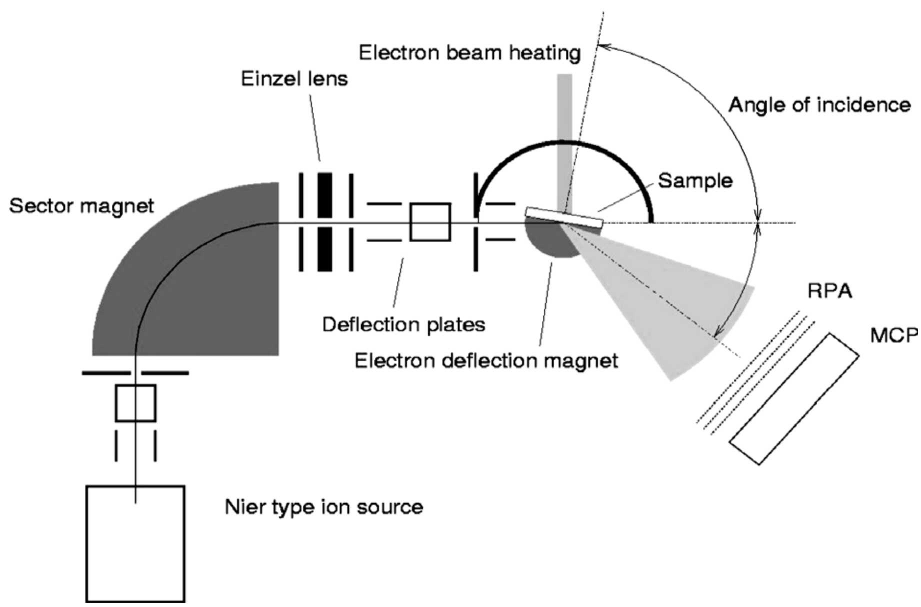

Fig. 1. Schematic representation of the ILENA experiment at the University of Bern. Positive ions are extracted from a Nier type ion source and after being mass analyzed scattered from a sample-surface under grazing angles of incidence. The distribution of scattered particles is recorded with an MCP-detector. An electron deflection magnet holds back electrons which are emitted during the scattering process. See text for further details.

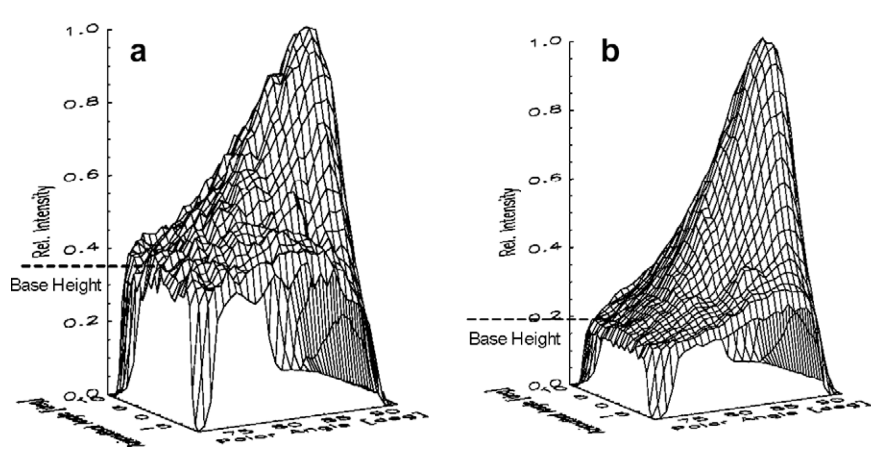

Fig. 2. Comparison of angular scattering distributions of 500 eV O_2^+ scattered from thin Al_2O_3 films with film thicknesses of 172 nm (a) and 25 nm (b). The angle of incidence is 82° with respect to the surface normal. The higher surface roughness of the thicker film causes a broader distribution of scattered particles, which is clearly seen.

The fraction of negative ions is determined by taking measurements with and without an applied floating voltage on the MCP. In the first case, only neutral particles are recorded, in the latter case neutral particles and negative ions are recorded simultaneously. The difference gives the fraction of negative ions. Each data-point results from a series of successive measurements, which allows the detection of possible ion beam instabilities and surface charging during each measurement series. For all of the thin films tested we could not find any sign of surface charging. With the MCP detector, we cannot distinguish between negatively ionized primary particles and sputtered negative ions. Therefore, all measurements were repeated with incident positive ions of comparable mass but without the ability to form stable negative ions (i.e. He for H_2, N for O) and the negative ion fractions recorded there were taken as sputtering background of the previous measurements. Subsequently, these sputtered fractions were subtracted from the measurements.

The detection efficiency of the MCP is taken from [19,20], where an identical detector was used.

3. Results and discussion

Although we eventually want to detect neutral atoms in space, we used positive molecular ions because they can be produced far more efficiently, and with much better energy, intensity, and angle control in our system than can neutrals. But the charge and mass of molecules must be justified.

From previous experiments with several other insulating surfaces (polycrystalline diamond [11], single-crystal diamond [21,22], and MgO [12]) it has been established that incident hydrogen and oxygen ions are effectively neutralized upon scattering. These previous measurements were done with both incident positive ions and with incident neutral particles and they revealed the same negative ion fractions in both cases. As a result, we can assume complete memory loss of the incident charge state after scattering.

The use of molecules instead of atoms is justified as follows. A molecule has many more electronic states than an atom, so we cannot expect the charge exchange process while scattering to be identical. But according to [10] and [11], more than 80% of molecules with energy in the 300–1000 eV range, when scattered off a polycrystalline diamond surface, dissociate shortly before reaching the surface, on the incoming stage of the trajectory. That means that the final charge state fraction is determined mainly by charge exchange processes between the surface and dissociated atoms. Therefore, we conclude that the use of molecules causes a negligible change to the charge state fractions measured in this study.

3.1. Angular scattering

The two key requirements for successful conversion surface performance in a space instrument are high ionization yield and low spread of scattering angles, the latter to minimize scattered particle loss in downstream detection systems. The component of angular deviation from specular scattering that resides in a plane containing the incoming trajectory and normal to the surface is defined as polar scattering. The component of specular scattering normal to the polar angle plane is defined as azimuthal scattering, with zero indicating a true specular reflection. Fig. 2 shows angular scattering distributions for two different thin Al_2O_3 films with film thicknesses of 172 nm (Fig. 2(a)) and 25 nm (Fig. 2(b)), respectively. In the latter case, the silicon substrate was etched with HF prior to the coating process, which may increase the smoothness of the film [23]. The observed asymmetric beam profile is caused by surface roughness, which inevitably leads to a certain fraction of scattered particles, which do not undergo specular reflection. Therefore, the beam profile of the scattered distribution is broadened, and this effect will increase with higher surface roughness, which is, as a first result, in this study equivalent to higher film thickness. There are two measures for angular scattering: FWHM in azimuthal and polar direction and base height (dashed line in Fig. 2) of the beam profiles. Fig. 3 displays these measures for 500 eV O_2^+ scattered from the thin Al_2O_3 films analyzed in this study in comparison to previous scattering experiments with MgO [12] and the surfaces, which are used as conversion surfaces for the IBEX-Lo instrument of NASA's IBEX mission, hydrogen terminated thin films of diamond-like carbon on silicon [9,10]. Of course, the film thickness of the IBEX surfaces is not zero, but they are included on Fig. 3 to serve as a reference. Obviously, the newly tested thin Al_2O_3 films are better (i.e. have narrower scattering cones and lower base height) than the previously tested MgO and Al_2O_3 surfaces, especially the 25 nm surface, which was etched with HF prior to the coating process. Although they are not as good as the IBEX surfaces, the differences are small enough to consider them for further testing.

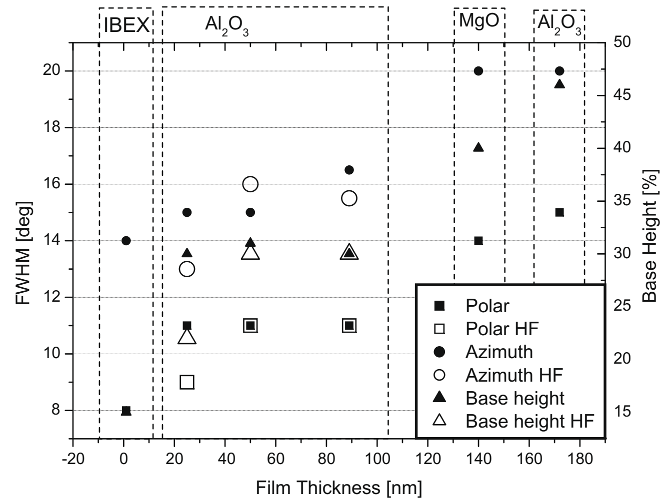

Fig. 3. FWHM of angular scattering versus film thickness of various thin films of Al_2O_3 on silicon substrates. Some substrates were etched with HF prior to the coating process with the goal of enhancing the film smoothness. The data for these surfaces is shown as "HF". The data is presented in comparison to previous measurements with MgO and the surfaces, which are used as conversion surfaces for the IBEX-Lo instrument of NASA's IBEX mission. Of course, the film thickness of these IBEX-Lo surfaces is not zero; they are included as a reference. All data points were taken with 500 eV O_2^+.

3.2. Ionization yield

Fig. 4 shows the negative ion fractions measured for scattering of oxygen and hydrogen ions from the different thin Al_2O_3 films. We found increasing negative ion fractions with increasing energy of the incident ions. Similar findings have been reported for other insulating surfaces such as LiF [8], MgO [12] and $BaZrO_3$ [24], and we follow the same argumentation, that higher energies cause smaller distances of closest approach between scattered ions and surface atoms, which results in higher probabilities for charge exchange processes and thus yields higher fractions of negative ions.

The negative ion fractions we recorded increased with energy for oxygen from 11% to 17% and for hydrogen from 1% to 2%. Within the measurement uncertainties of ±1% for oxygen and ±0.5% for hydrogen all films show similar ion fractions. However, a close look reveals that for the smoothest surface, i.e. 25 nm with HF etching, the negative ion fractions are slightly lower, which is in good

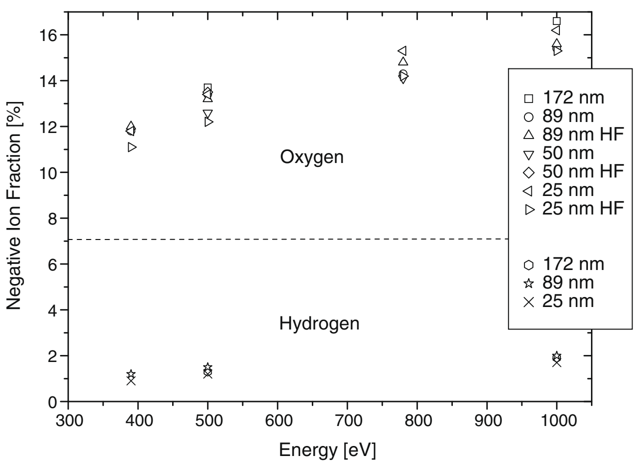

Fig. 4. Energy dependence of negative ion fractions resulting from positive molecular oxygen and hydrogen ions scattered from various thin films of Al_2O_3 on silicon substrates. To enhance the film smoothness some of the substrates were etched with HF prior to the coating process. Results for these films are indicated as "HF".

agreement with previous findings [9,10,16], that higher surface roughness increases the probability for violent collisions and thus the probability for charge exchange processes.

Regarding the negative electron affinities of oxygen (1.46 eV) and hydrogen (0.75 eV), one should expect for higher negative electron affinities higher negative ion fractions, which explains the higher negative ion yield for oxygen than for hydrogen seen in Fig. 4.

It has yet to be explained how charge exchange happens when particles scatter off an insulating surface. According to [25] and [26], the width of the band gap of Al_2O_3 is about 9 eV. As mentioned before, the negative electron affinity levels of H and O are 0.75 and 1.46 eV below the vacuum level, respectively. The valence band is filled and there is no electron mobility in an insulator. So one should not expect that charge exchange would be possible. But it has been found that for ionic crystals such as, for instance, LiF, charge exchange proceeds via capture of electrons from the anionic sites of the surface in a binary ion–atom interaction [27–29]. And once the negative ion is formed it cannot be destroyed by resonant electron loss (as in the case of metals) because of the band gap of the ionic crystal. It is safe to expect a similar behaviour here. In addition, the probability for a particle to be negatively charged increases with increasing effective number of collisions, and thus at grazing incidence angles.

4. Conclusions

The motivation for this study was the question of whether thin Al_2O_3 films would be an alternative to work as conversion surfaces in neutral particle sensing instruments. Taking both angular scattering and ionization yield into account, we have to admit that the surfaces we use for the IBEX-Lo instrument of NASA's IBEX mission [10] are still the best surfaces we found so far. However, if price and especially availability are crucial, smooth thin Al_2O_3 films such as the tested 25 nm thick Al_2O_3 film on a HF etched silicon substrate could indeed be an alterative. Equipped with these surfaces, an instrument like IBEX-Lo would be less efficient because of lower ionization yield and larger angular scattering (and thus lower overall transmission of the instrument). On the other hand, lower transmission would be acceptable if the scientific requirements (depending on expected particle flux, for instance) fit the efficiency of this instrument.

Acknowledgements

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References

[1] C. Auth, A.G. Borisov, H. Winter, Phys. Rev. Lett. 75 (1995) 2292.
[2] P. Stracke, F. Wiegershaus, S. Krischok, H. Müller, V. Kempter, Nucl. Instr. and Meth. B 125 (1997) 63.
[3] S.A. Deutscher, A.G. Borisov, V. Sidis, Phys. Rev. A 59 (1999) 4446.
[4] P. Roncin, J. Villette, J.P. Atanas, H. Khemliche, Phys. Rev. Lett. 83 (1999) 864.
[5] A.G. Borisov, V.A. Esaulov, J. Phys.: Condens. Matter 12 (2000) R177.
[6] R. Souda, Int. J. Mod. Phys. B 14 (2000) 1139.
[7] H. Winter, Phys. Rep. 367 (2002) 387.
[8] J.A. Scheer, P. Wurz, W. Heiland, Nucl. Instr. and Meth. B 212 (2003) 291.
[9] J.A. Scheer, M. Wieser, P. Wurz, P. Bochsler, E. Hertzberg, S.A. Fuselier, F.A. Koeck, R.J. Nemanich, M. Schleberger, Nucl. Instr. and Meth. B 230 (2005) 330.
[10] J.A. Scheer, M. Wieser, P. Wurz, P. Bochsler, E. Hertzberg, S.A. Fuselier, F.A. Koeck, R.J. Nemanich, M. Schleberger, Adv. Space Res. 38 (2006) 664.
[11] P. Wurz, R. Schletti, M.R. Aellig, Surf. Sci. 373 (1997) 56.
[12] M. Wieser, P. Wurz, K. Brüning, W. Heiland, Nucl. Instr. and Meth. B 192 (2002) 370.
[13] P. Wurz, Detection of Energetic Neutral Particles, in: The Outer Heliosphere: Beyond the Planets, Copernikus Gesellschaft e.V., Katlenburg-Lindau, 2000, p. 251.
[14] T.E. Moore, D.J. Chornay, M.R. Collier, F.A. Herrero, J. Johnson, M.A. Johnson, J.W. Keller, J.F. Laudadio, J.F. Lobell, K.W. Ogilvie, P. Rozmarynowski, S.A. Fuselier, A.G. Ghielmetti, E. Hertzberg, D.C. Hamilton, R. Lundgren, P. Wilson, P. Walpole, T.M. Stephen, B.L. Peko, B. van Zyl, P. Wurz, J.M. Quinn, G.R. Wilson, Space Sci. Rev. 91 (2000) 155.
[15] P. Wurz, M.R. Aellig, P. Bochsler, A.G. Ghielmetti, E.G. Shelley, S. Fuselier, F. Herrero, M.F. Smith, T. Stephen, Opt. Eng. 34 (1995) 2365.
[16] J.A. Scheer, P. Wahlstroem, P. Wurz, Nucl. Instr. and Meth. B 256 (2007) 76.
[17] J. Geiss, M. Witte, Space Sci. Rev. 78 (1996) 229.
[18] S. Jans, P. Wurz, R. Schletti, T. Fröhlich, E. Hertzberg, S. Fuselier, J. Appl. Phys. 87 (2000) 2587.
[19] B.L. Peko, T.M. Stephen, Nucl. Instr. and Meth. B 171 (2000) 597.
[20] T.M. Stephen, B.L. Peko, Rev. Sci. Instr. 71 (2000) 1355.
[21] J.A. Scheer, K. Brüning, T. Fröhlich, P. Wurz, W. Heiland, Nucl. Instr. and Meth. B 157 (1999) 208.
[22] P. Wurz, T. Fröhlich, K. Brüning, J.A. Scheer, W. Heiland, E. Hertzberg, S. Fuselier, in: J. Safrankov, A. Kanka (Eds.), Prague Proceeding Week of Postdoc. Students, 1998, p. 257.
[23] H. Schlemm, JENION, personal communication.
[24] S. Jans, P. Wurz, R. Schletti, K. Brüning, K. Sekar, W. Heiland, J. Quinn, R.E. Leuchter, Nucl. Instr. and Meth. B 173 (2001) 503.
[25] I. Batra, J. Phys. C: Solid State Phys. 15 (1982) 5399.
[26] V.M. Fomin, V.R. Misko, J.T. Devreese, H.H. Brongersma, Nucl. Instr. and Meth. B 145 (1998) 545.
[27] C. Auth, A. Mertens, H. Winter, A.G. Borisov, V. Sidis, Phys. Rev. A 57 (1998) 351.
[28] F.W. Meyer, Q. Yan, P. Zeijlmans van Emmichoven, I.G. Hughes, G. Spierings, Nucl. Instr. and Meth. B 125 (1997) 138.
[29] S. Ustaze, R. Verrucchi, S. Lacombe, L. Guillemot, V.A. Esaulov, Phys. Rev. Lett. 79 (1997) 3526.