Available online at www.sciencedirect.com
SCIENCE DIRECT®
Nuclear Instruments and Methods in Physics Research B 212 (2003) 291–296
NIM B Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

Scattering of slow ions from insulator surfaces at the example of molecular oxygen from LiF(1 0 0)

J.A. Scheer [a,*], P. Wurz [a], W. Heiland [b]

[a] *Phys. Institut, Universität Bern, CH-3012 Bern, Switzerland*
[b] *Universität Osnabrück, D-49069 Osnabrück, Germany*

Abstract

We scattered molecular oxygen ions at grazing incidence from a LiF(1 0 0) surface in the energy range from 880 to 3040 eV. About 50% molecular survival was obtained at the lowest primary energy of 881 eV and this dropped to about 4% at 3039 eV. Below 2 keV beam energy we found negative ion fractions of less than 2%, which increases up to 6% at about 3 keV primary energy. The molecular survival is much higher whereas the negative ion fraction is much lower compared to other insulating materials, i.e. diamond and MgO. The mean energy loss is found to increase linearly with energy and is in the order of a few percent.

PACS: 78.70.−g; 61.18.Bn
Keywords: LiF; Neutral particle imaging; Particle scattering; Surface ionization

---

1. Introduction

Interaction of atomic and molecular projectiles with insulating surfaces of ionic crystals has gained a lot of attention in recent years [1–7]. Reports about the relative high fractions of negative ions while scattering atomic and molecular ions from insulating surfaces came quite unexpected but gave ideas for several applications. We want to use this process for the efficient detection of energetic neutral particles in interplanetary and interstellar space [8–10]. The mass spectrograph on the IMAGE satellite mission, designed to detect low energy neutral atoms, uses a conversion surface of volatile adsorbates on a polished W substrate to convert a fraction of the incoming neutral atoms to negatively charged ions [8,11]. For future missions, a conversion surface with better long-term stability and without the need of continuous preparation is desired. The understanding of the interaction process between ions and insulating surfaces is crucial to find a suitable surface. It is expected that the neutral components of interstellar gas consist mainly of H, He and O. We scattered molecular oxygen ions from LiF(1 0 0) which is an ionic conductor material with a very wide bandgap of 14 eV and compare the results with previous measurements, for instance molecular hydrogen on LiF(1 0 0) [12] and molecular oxygen on MgO(1 0 0) [9].

---

[*] Corresponding author. Fax: +41-31-631-4405.
E-mail address: jscheer@phim.unibe.ch (J.A. Scheer).

0168-583X/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/S0168-583X(03)01838-X

2. Experiment

The experimental setup has been described elsewhere in detail [13]. The apparatus consists of a UHV chamber coupled to a low-energy ion accelerator system. The accelerator system consists of a plasma ion source where any gaseous ions can be produced. The charged gaseous ions are then energy and mass filtered by a magnet and the beam is directed onto the sample placed in a UHV system. A gas cell present between the analyzing magnet and the target chamber is used to produce neutral beams if neutral incident particles are used. The base pressure in the target chamber is about 3×10^{-10} mbar, during the measurements it rises up to about 1×10^{-9} mbar. The target is mounted on a three-axis manipulator equipped with electron beam heating. The temperature is controlled by a thermocouple. For our experiments with LiF(1 0 0) we use a target holder where we can mount two targets in parallel as we already reported earlier [14]. The targets are aligned with respect to the vertical movement of the manipulator. That has the advantage that all beam adjustments could be made using a metal target, here a W single crystal, without causing possible damage to the LiF(1 0 0) sample or having problems with charging. Furthermore, from earlier experiments [12] we learnt that at temperatures above 250 °C LiF is a good ionic conductor. Therefore, all measurements have been done at a sample temperature of 300 °C to avoid pileup of surface charges that otherwise will deflect the incident ion trajectory. The energy of the scattered particles is measured using a time-of-flight (TOF) system. The TOF-beamline is fixed at an scattering angle of 10° with respect to the primary beam direction and all measurements are performed under specular reflection, i.e. a glancing angle of 5° is used. Particle detection is achieved by using a multichannel plate detector (MCP) with an aperture of 1.2° (full width). By using an appropriate postacceleration voltage separation of neutral and different charged particles could be achieved.

The LiF(1 0 0) surface was sputtered slightly with 1 keV Ar^+ at angles of incidence between 5° and 15°. The target cleanliness was probed by the TOF technique looking for recoil ions. Estel et al. [15] showed earlier that the LiF surface is very stable. At room temperature they showed no adsorption on LiF when they tried to adsorb water.

3. Results and discussion

The measurements have been performed with O_2^+ incident on a LiF(1 0 0) surface along an arbitrarily chosen direction, i.e. the sample has not been aligned to any crystallographic direction, with primary energies varying from 881 eV up to 3039 eV. Fig. 1 shows two spectra for different

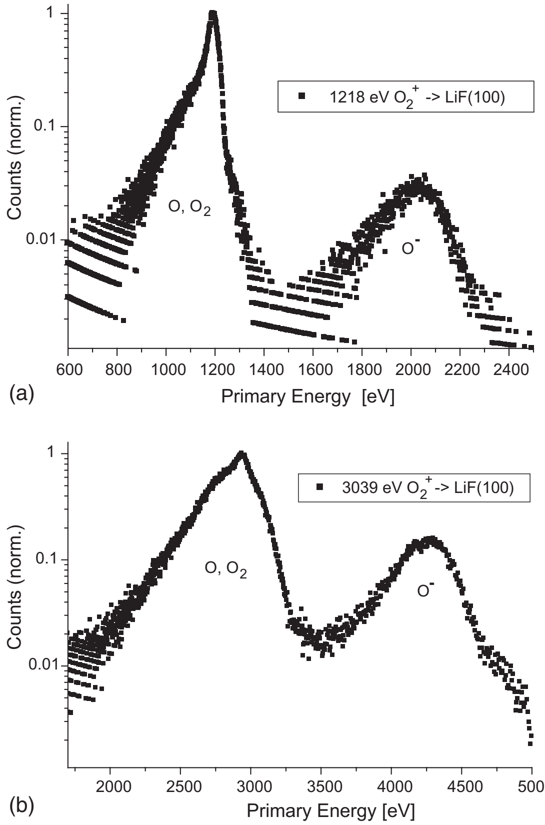

Fig. 1. Energy spectra of O, O_2 and O^- for O_2^+ incident on LiF(1 0 0) at an angle of incidence of 5° for various primary energies. The scattering angle is 10°. Each spectrum shows a superposition of neutral O and O_2 followed by an additional peak of O^- at higher energies due to an applied postacceleration voltage. The superposition consists of a narrow peak of surviving molecules riding over a broad distribution of atoms from dissociated molecules.

primary energies. The spectra have been transformed from time-of-flight spectra into energy spectra. The distribution of the neutral particles (O, O_2) is on the lower energy side. The surviving molecules form a narrow peak that rides over the broad peak of the atoms from dissociated molecules. Because of the dissociation energy released during the dissociation process the high-energy tail of the atomic peak exceeds the primary energy. On the right hand side, at higher energies, a negative atomic oxygen ion (O^-) peak is seen. It has been separated from the neutral peaks by applying a postacceleration voltage. All energy spectra have been taken with positive and negative postacceleration voltage. Positive oxygen ions could be detected only for the highest primary energies investigated and their fraction was less than 1%. Furthermore, we never saw any negative molecular oxygen ions (O_2^-), which differs from previous measurements on palladium or diamond [16].

The data were corrected for the relative detection efficiency of the MCP detector and then analyzed as described in [17,18]. In addition, to fit the atomic peaks a so-called exponentially modified Gauss function (EMG) has been used, which is basically a convolution of a gaussian peak with an exponential function as proposed, for instance, in [19]. The exponential part takes inelastic energy losses of the particles into account, i.e. the lower energy tails of the peaks (straggling). For molecules, this effect has been neglected, which is supported by the energy loss we could detect for molecules (Fig. 4); therefore a pure gaussian function has been used. This is of course an idealization but allows one to fit even spectra where the atomic and the molecular peak are not so distinct as in Fig. 1(a). Overall, the fitted distributions match very well with the experimental data.

Already from the "raw data" presented in Fig. 1, comparing the narrow peaks of the molecules in Fig. 1(a) and (b), it is clear that for higher primary energies the survival probability of molecules decreases remarkably. Fig. 2 shows the molecular survival fraction as a function of primary energy. From about 50% at 1 keV the fraction drops down below 4% at 3 keV. Compared to a metal surface this is quite a high survival fraction, for example especially for low energies it is more than twice as much as for scattering of O_2^+ from a palladium surface [13]. This higher dissociation on metal surfaces is caused by electron capture into an antibonding triplet state [20], which is comparably unlikely on a wide bandgap insulator.

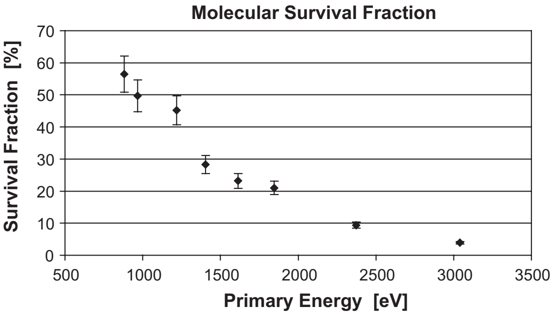

Fig. 2. Molecular survival fraction of O_2 scattered from a LiF(1 0 0) surface at 5° incidence angle and 10° scattering angle.

Furthermore, while scattering N_2^+ from a palladium surface survival fractions have been found which were even higher, i.e. about 70% at 1 keV. From previous experiments we know that N_2 dissociates when scattered off metal surfaces by vibrational and rotational excitation only, which means that there is no charge exchange involved in the dissociation process [14]. Taken that into account, and also that the binding energy for N_2 (226 kcal/mol) is about a factor of two larger than the binding energy for O_2 (118 kcal/mol) which results in a higher survival probability for N_2 compared to O_2, one can conclude that the dissociation process for O_2^+ scattered off LiF is also largely driven by mechanical excitations of the molecule. Dissociation via charge exchange cannot be excluded but is surely not the dominating channel.

Compared to other insulator surfaces there are quite different results. While scattering of molecules off a diamond surface shows a similar behavior (though lower yields of surviving molecules) scattering experiments with magnesium oxide (MgO) [9] and with barium zirconate ($BaZrO_3$) [21] surfaces within the same primary energy range revealed no surviving molecules at all. An easy explanation would be a remaining surface contamination in the two latter cases, i.e. an oxide layer, but there might be other reasons as well.

For O_2^+ scattered off LiF(1 0 0) Fig. 3 displays the charge state fraction of O^- as a function of primary energy. The relatively high values at the two lowest energies are probably due to poor quality of the spectra taken at primary energies below 1 keV causing difficulties for the fitting procedure. However, since these values were more or less reproducible they are shown here. And for the molecular survival fraction this uncertainty does not play an important role. The negative ion fraction rises from less than 1% at 1 keV to about 6% at 3 keV primary energy. The increase of the negative ion fraction with increasing primary energy could already be derived from the comparison of Fig. 1(a) and (b). This rise has also been observed at scattering experiments with molecular hydrogen off LiF(1 0 0) [12] and is also predicted by theory [22,23]. Furthermore, the negative ion fractions found at MgO [9], $BaZrO_3$ [21] and aluminum-nitride (AlN) [24] show a similar tendency but with values of 15–30%. Winter et al. [25] reported negative ion fractions up to 60% while scattering oxygen ions from LiF(1 0 0) but these experiments were at much higher primary energies and for an angle of incidence of about 1°. On the other hand, at the low energy part their fractions drop below 10%, which fit quite nicely to our data. The charge capture from LiF is proposed to be of a binary type of collisions between the anionic site, F^-, and the incident ion. Once the negative ion is produced, due to the large bandgap of LiF (14 eV) subsequent electron loss is not possible for the negative ion in contrast to the situation on a metal surface [1,22,25], with the possible exception of surface states [26].

In Fig. 4 we present the measured energy losses for dissociated atoms and surviving molecules as a function of primary energy per ion. It was not possible to get unambiguous values for the two lowest energies so that, for clarity reasons, they are not shown here and not considered in the following discussion. For molecules as well as for atoms the energy loss increases linearly with incident energy. This is in good agreement with results reported earlier [4,9,21,24,27]. For scattering of hydrogen there are reports that at low primary energies the energy loss becomes almost constant [12,28]. We did not observe this, maybe because of the limits of our experimental setup. Compared with the primary energy the energy loss of the surviving molecules is about 2%. In general, up to 1% is attributed to elastic scattering and the rest to inelastic effects, i.e. charge exchange, straggling and so on. This result supports the concept that the inelastic energy loss of a molecule has to be very low because otherwise it will dissociate due to electrical or mechanical excitations. Consequently, for atoms we measured a relative energy loss of more than 4%. This is in the region of hydrogen measurements with LiF(1 0 0) (3–7%) [12], but it is

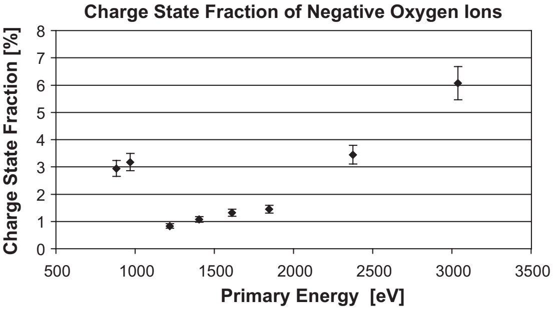

Fig. 3. Charge state fraction for O^- at various O_2^+ primary energies scattered off a LiF(1 0 0) surface. No molecular negative ions could be detected. Positive ions were visible at the highest primary energies but their fraction was well below 1%, i.e. their distribution was hardly to separate from the background. Note that the relative high charge state fraction at the two lowest primary energies might be caused by the poor quality of spectra taken at primary energies below 1 keV which increases the uncertainty of the fitting procedure.

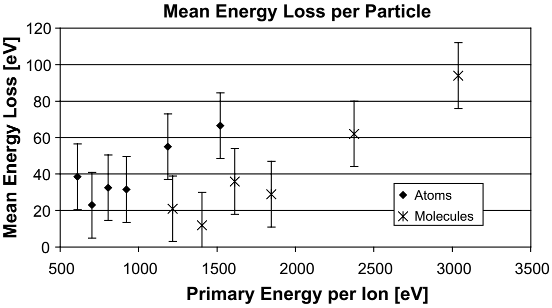

Fig. 4. Energy loss of neutral oxygen ions at various primary energies per ion. The relative energy loss (compared to the primary energy) is for molecules about 2% and for atoms about 4%.

very low compared to the scattering of oxygen from MgO [9] and from $BaZrO_3$ [21] where energy losses of 12–20% have been found. However, the results for MgO and $BaZrO_3$ are also rather high compared to results reported in literature for other particles scattered from metallic surfaces [7]. On the other hand, recent studies showed a "metal-like" behavior of stopping at surfaces for ionic crystals like LiF [29], which means in short that there is no threshold effect for projectile stopping with respect to ion energy or velocity caused by the energy gap. This agrees pretty well with our measurements. The authors argue that this "metal-like" behavior appears when projectile and target approach so close that molecular orbitals are formed between projectile and target which lead to electron promotion and finally to a reduction of energy defects in comparison to the unperturbed energy gap of the crystal. As necessary minimum energy for this effect to take place they report 2 keV/amu. This is far more than the primary energies we used ($\sim$1–3 keV), but their measurements have been done with an incident angle of about 1°, so if we compare the components of the ion velocities perpendicular to the surface we find very similar results and can therefore assume similar distances of closest approach between projectile and target.

4. Conclusion

Scattering of O_2^+ molecules from a LiF(1 0 0) surface at grazing incidence has been studied in the primary energy range 881–3039 eV. We found large fractions of surviving molecules after scattering, which dropped from about 50% at 1 keV down to about 4% at 3 keV. The mean energy loss of the dissociated atoms increases linearly and is of the order of 4%, which agrees well with existing theories. Within the energy range observed the negative ion fraction increases with primary energy but is, at least compared with hydrogen molecules, quite low, i.e. less than 2% below 2 keV primary energy. This is much lower than for diamond, AlN, $BaZrO_3$ and MgO [9,14,21,24]. It appears that there is a correlation between ion fraction and secondary electron emission. LiF has one of the lowest secondary electron emission coefficients of the insulators, MgO the highest [30].

Acknowledgements

This work is supported by the Deutsche Forschungsgemeinschaft and the Swiss National Science Foundation.

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] P. Wurz, R. Schletti, M.R. Aellig, Surf. Sci 373 (1997) 56.

[9] M. Wieser, P. Wurz, K. Brüning, W. Heiland, Nucl. Instr. and Meth. B 192 (2002) 370.

[10] P. Wurz, Detection of energetic neutral particles, in: The Outer Heliosphere: Beyond the Planets, Copernikus Gesellschaft e.V., Katlenburg-Lindau, 2000, p. 251.

[11] T.E. Moore et al., Geophys. Res. Lett. 28 (6) (2001) 1143.

[12] K. Sekar, J.A. Scheer, K. Brüning, W. Heiland, Phys. Rev. B 63 (2001) 045411.

[13] B. Willerding, H. Steininger, K.J. Snowdon, W. Heiland, Nucl. Instr. and Meth. B 2 (1984) 453.

[14] J.A. Scheer, K. Brüning, T. Fröhlich, P. Wurz, W. Heiland, Nucl. Instr. and Meth. B 157 (1999) 208.

[15] J. Estel, H. Hoinkes, H. Kaarmann, H. Nahr, H. Wilsch, Surf. Sci. 54 (1976) 393.

[16] J.A. Scheer, Master Thesis, University of Osnabrück, 1999.

[17] K. Schmidt, H. Franke, T. Schlathölter, C. Höfner, A. Närmann, W. Heiland, Surf. Sci. 301 (1994) 326.

[18] W. Heiland, T. Schlathölter, M. Vicanek, Phys. Status Solidi B 192 (1995) 301.

[19] A. Närmann, Dissertation Thesis, University of Osnabrück, 1989.

[20] W. Heiland, The interaction of molecular ions with surfaces, in: J.W. Rabalais (Ed.), Low Energy Ion-Surface Interactions, John Wiley and Sons Ltd., 1994, p. 313.

[21] 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.

[22] A.G. Borisov, V. Sidis, Phys. Rev. B 56 (1997) 10628.

[23] N. Lorente, J. Merino, F. Flores, M.Y. Gusev, Nucl. Instr. and Meth. B 125 (1997) 77.

[24] S. Jans, P. Wurz, R. Schletti, T. Fröhlich, E. Hertzberg, S. Fuselier, J. Appl. Phys. 87 (2000) 2587.

[25] H. Winter, C. Auth, A.G. Borisov, Nucl. Instr. and Meth. B 115 (1996) 133.

[26] P. Wurz, J. Sarnthein, W. Husinsky, G. Betz, P. Nordlander, Y. Wang, Phys. Rev. B 43 (1991) 6729.

[27] C. Auth, A. Mertens, H. Winter, A. Borisov, Phys. Rev. Lett. 81 (1998) 4831.

[28] H. Winter, C. Auth, A. Mertens, Nucl. Instr. and Meth. B 164–165 (2000) 559.

[29] J.I. Juaristi, C. Auth, H. Winter, A. Arnau, K. Eder, D. Semrad, F. Aumayr, P. Bauer, P.M. Echenique, Phys. Rev. Lett. 84 (2000) 2124.

[30] J. Cazaux, Some physical descriptions of the charging effects in insulators under irradiation, in: Ionization of Solids by Heavy Particles, Plenum Press, New York, 1993, p. 325.