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<p>The intent of this research program was to elucidate the microscopic mechanisms by which the energy of incoming beams of atoms, ions, electrons and photons is absorbed, localized and redirected to produce neutral-particle desorption from surfaces and subsequent surface modification or restructuring. The model system LiF was used, and the results compared with measurements of electronically desorbed Li-atoms adsorbed on other substrates. This study explored how the degree of surface metallization influences the choice of the final excitation state of the desorbing particle. Measurements were made to compare the desorption of excited state neutral lithium from lithium fluoride by electron, photon and ion bombardment and from lithium-dosed tungsten and lithium-dosed glass by electron and photon bombardment. In the electron and photon desorption experiments, we observed only the first lithium resonance line. These results contrast with the ion sputtering results, where emissions from higher excited states were observed. This suggests that the presence of a metal rich surface provides a channel for de-excitation of excited lithium states which are above the lithium metal Fermi energy.</p>			
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COMPLETED PROJECT SUMMARY

TITLE: Electronic Interactions of Electrons, Photons and Atoms with Material Surfaces

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PUBLICATIONS:

"Energy Shifts and Broadening of Excited Hydrogen-Atom Levels in the Vicinity of a Metal Surface," P. Nordlander and J. C. Tully, Phys. Rev. Letters **61**, 990 (1988)

"Electronic Transitions in Surface and Near-Surface Radiation Effects," R. F. Haglund, Jr., M. H. Mendenhall, N. H. Tolk, G. Betz, and W. Husinsky, Nucl. Instr. and Meth. **B32**, 321 (1988)

"Extended Huckel Theory for Ionic Molecules and Solids: An Application to Alkali Halides," Y. Wang, P. Nordlander, and N. H. Tolk, to be published in the October 1988 issue of J. Chem. Phys.

"Ultraviolet Spectroscopy of CN⁻ in Alkali Halides: Dynamics of the Metastable Triplet State," M. Mendenhall, A. Barnes, P. Bunton, R. Haglund, L. Hudson, R. Rosenberg, D. Russell, J. Sarnthein, P. Savundararaj, N. Tolk, and J. Tellinghuisen, Chem. Phys. Letters **147**, 59 (1988)

"PSD of Excited Hydrogen from KCl," L. T. Hudson, A. V. Barnes, N. J. Halas, R. F. Haglund, M. H. Mendenhall, P. Nordlander, N. H. Tolk, Y. Wang, and R. A. Rosenberg, in Desorption Induced by Electronic Transitions, DIET III ed. by R.H. Stulen and M.L. Knotek (Springer-Verlag, Berlin, 1988) p. 274

"Optical Radiation from Electron, Photon and Heavy Particle Bombardment of Lithium Fluoride and Lithium-Dosed Surfaces," N. H. Tolk, R. G. Albridge, A. V. Barnes, R. F. Haglund, Jr., L. T. Hudson, M. H. Mendenhall, D. P. Russell, J. Sarnthein, P. M. Savundararaj, and P. W. Wang, in Desorption Induced by Electronic Transitions, DIET III, ed. by R.H. Stulen and M.L. Knotek (Springer-Verlag, Berlin, 1988) p. 284

"Long Lifetime of Bulk Luminescence Observed in Spectrosil Glasses Under Electron Bombardment," P. W. Wang, R. F. Haglund, Jr., L. T. Hudson, D. L. Kinser, M. H. Mendenhall, N. H. Tolk, and R. A. Weeks, in Desorption Induced by Electronic Transitions, DIET III, ed. by R.H. Stulen and M.L. Knotek (Springer-Verlag, Berlin, 1988) p. 289

"Intracavity Optical Damage Due to Electrons, Ions and Ultraviolet Photons," R. F. Haglund, Jr., SPIE 895, 182 (1988)

ABSTRACT OF OBJECTIVES AND ACCOMPLISHMENTS:

The intent of this research program is to elucidate the microscopic mechanisms by which the energy of incoming beams of atoms, ions, electrons and photons is absorbed, localized and redirected to produce neutral-particle desorption from surfaces and subsequent surface modification or restructuring. In particular, we emphasized (1) heavy-particle-, electron-, and photon-induced desorption measurements for the case of the model system LiF, (2) kinematically-complete experiments in which all the neutral species desorbed from a surface are detected, in order to form a complete picture of the energy balance in electronically induced desorption processes for the LiF model system, (3) Parametric and time resolved studies of electronically-stimulated desorption as a function of incident-beam energy, beam intensity, and substrate conditions, and (4) comparison of the results of the above experiments with measurements of electronically desorbed Li-atoms coadsorbed with O-atoms on well defined crystals.

These comparative studies of electron, photon and ion bombardment of the same sample surface probed the various channels through which incident particle energy is dissipated. Electron or photon irradiation of alkali halides results in the swift ejection of halide atoms, leaving behind an enriched alkali metal surface from which the alkali atoms thermally desorb. Ion bombardment involves momentum transfer as well as electronic mechanisms, which results in a different surface stoichiometry at the time of desorption. This study explored how the degree of surface metallization influences the choice of the final excitation state of the desorbing particle. Measurements were made to compare the desorption of excited state neutral lithium from lithium fluoride by electron, photon and ion bombardment and from lithium-dosed tungsten and lithium-dosed glass by electron and photon bombardment.

In the electron and photon desorption experiments, we observed only the first lithium resonance line. These results contrast with the ion sputtering results, where emissions from higher excited states are observed. This suggests that the presence of a metal rich surface provides a channel for de-excitation of excited lithium states which are above the lithium metal Fermi energy. This quenching of the higher excited lithium lines is less likely when the sample is excited by ions. These studies have shown substantial success and have pointed the way for significant future progress. Further studies presently supported by AFOSR on another contract involve careful surface analysis of the stoichiometry of lithium fluoride during ion, electron and photon irradiation. The ultimate aim of this research program is to characterize the final states of all the desorption products and the extent to which they are influenced by the surface, exciting beam and secondary processes.

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**Electronic Interactions of Electrons, Photons, and Atoms
with Material Surfaces**

Grant AFOSR 86-0150

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Final Report for the period 15 April 1986 - 14 April 1988

Prepared for

*Air Force Office of Scientific Research
Directorate of Chemical and Atmospheric Sciences
Department of the Air Force
Bolling Air Force Base, DC 20332-6448*

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1. Research Objectives

The object of this research program was to address specific questions related to the ways in which energy is absorbed, localized and redistributed in electronically-stimulated desorption processes. Thus an experimental study was carried out of the various energy-transfer channels through which bombardment by energetic electron, heavy-particle, and photon beams leads to (a) surface modification, erosion and macroscopic damage of the LiF model system, and (b) desorption of Li atoms adsorbed on well defined surfaces. In particular, we emphasized (1) heavy-particle-, electron-, and photon-induced desorption measurements for the case of the model system LiF, (2) kinematically-complete experiments in which all the neutral species desorbed from a surface are detected, in order to form a complete picture of the energy balance in electronically induced desorption processes for the LiF model system, (3) parametric and time resolved studies of electronically-stimulated desorption as a function of incident-beam energy, beam intensity, and substrate conditions, and (4) comparison of the results of the above experiments with measurements of electronically desorbed Li-atoms coadsorbed with O-atoms on well defined surfaces.

This research bore directly on a broad spectrum of insulating materials involving considerations of time resolved and spatial energy localization measurements, exciton creation, and the role of defects at surfaces and at interfaces in altering local potentials which are involved in bond-making and breaking processes. In previous studies, we have established that neutral particle desorption via electronic transitions is a dominant effect of low-energy photon, electron and ion irradiation incident on many insulating surfaces, and plays a pivotal role in surface modification and damage in such materials¹. The mechanisms by which this desorption occurs are even yet only partially understood. However, it is clear that *desorption induced by electronic transitions* (DIET) is an ubiquitous phenomenon, occurring on the surface of every material thus far studied. The desired end result of this research program was the identification and elucidation of the primary mechanisms through which the deposition of electronic energy in the near surface bulk leads to desorption and consequently to changes in surface structure and composition. Significant progress was made in carrying out the research objectives.

The experiments performed in the program were concerned with the most basic questions of surface physics and chemistry, involving the geometrical structure, electronic structure and dynamics of bond-making and -breaking at surfaces. These comparative studies of electron, photon and ion bombardment of the same sample surface probed the various channels through which incident

particle energy is dissipated. Electron or photon irradiation of alkali halides results in the swift ejection of halide atoms, leaving behind an enriched alkali metal surface from which the alkali atoms thermally desorb. Ion bombardment involves momentum transfer as well as electronic mechanisms, which results in a different surface stoichiometry at the time of desorption. This study explored how the degree of surface metallization influences the choice of the final excitation state of the desorbing particle. Measurements were made to compare the desorption of excited state neutral lithium from lithium fluoride by electron, photon and ion bombardment and from lithium-dosed tungsten and lithium-dosed glass by electron and photon bombardment.

2. Research Status

A. PSD of Li* from Lithium Fluoride

Photon irradiation studies were performed at the University of Wisconsin's Synchrotron Radiation Center at Stoughton, Wisconsin. Zero order visible and ultraviolet photons from the two meter grazing incidence Brown-Lien-Pruett monochromator (20 - 1200 eV) were incident along the surface normal upon a LiF crystal which was previously cleaved in air and baked under UHV conditions at 600°C. The photon detection system included a 0.3 m McPherson 218 monochromator which has a resolution of 26.5 Å at 1 mm slit width, a photo-multiplier in a cooled housing, and a multi-channel analyzer or CAMAC crate for data acquisition. We scanned the spectral region of 2000-7000 Å using a 1200 lines/mm grating blazed at 5000 Å. Fluorescence radiation was measured at 90° to the surface normal. The base pressure in the experimental chamber was less than 3×10^{-10} Torr throughout these experiments.

The spectrum obtained from irradiation of lithium fluoride (Fig.1) at room temperature shows the first resonance line of lithium at 6708 Å and two broad continua centered at about 3200 Å and 5600 Å. These continua are due to bulk luminescence and scattered light from the irradiating zero order photon beam. No other line radiation was detected above the background noise. We estimate that other spectral lines have maximum intensities that are at least a factor of 100 less intense than the intensity of the observed resonance line.

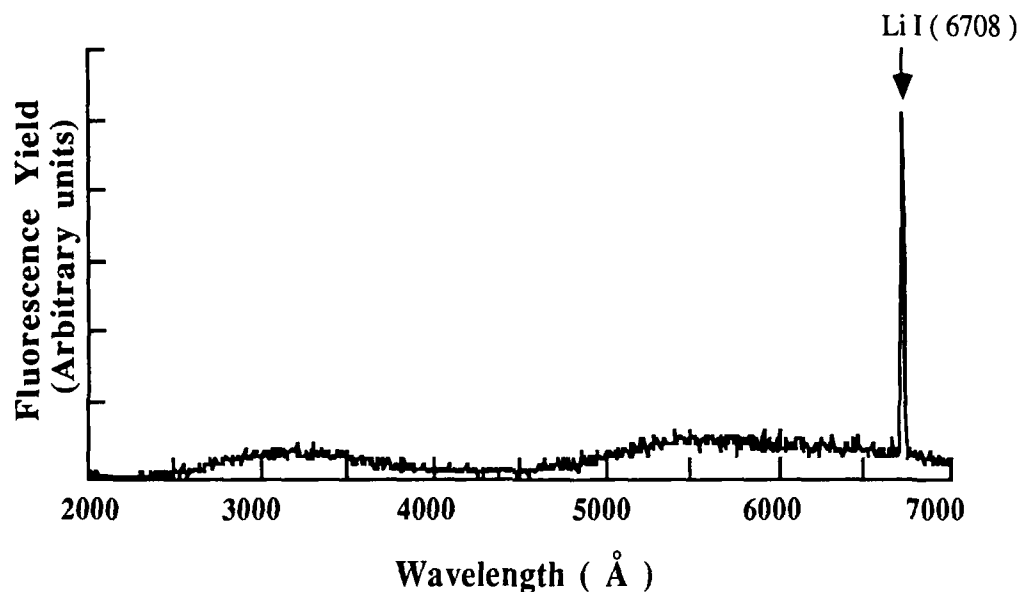


Fig.1 Fluorescence spectrum from photon irradiation of lithium fluoride

B. Ion Bombardment of Lithium Fluoride

In further studies, we irradiated a similarly prepared lithium fluoride sample with a 9 keV H_2^+ beam from a Colutron ion source. The de-excitation of the desorbed species from the surface and the bulk luminescence were monitored by the signal detection scheme discussed in part A. The ion bombardment was performed at room temperature. Note that in addition to the first resonance line of lithium 6708 Å (2p-2s) there are lithium lines at 3233 Å (3p-2s), 4603 Å (4d-2p) and 6104 Å (3d-2p). The relative intensities of the excitations originating at $n \geq 3$ after correcting for detection system efficiency are greatly enhanced compared to arc discharge data. This indicates a preferential population of these higher excited states via some unknown mechanism.

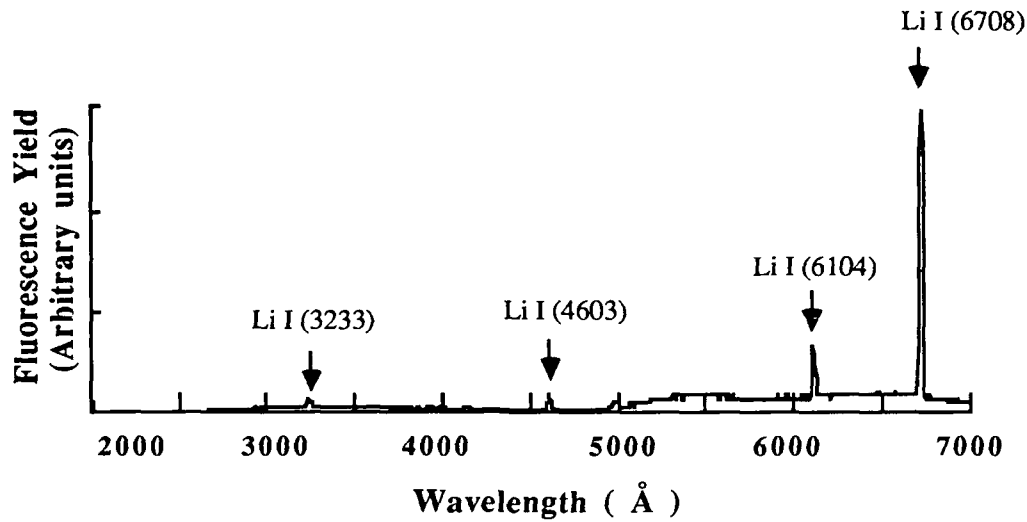


Fig 2. Fluorescence spectrum from 9 keV H_2^+ bombardment of lithium fluoride

C. ESD of Li^* from Lithium Fluoride

It is well known that electron bombardment of alkali halides can desorb generous amounts of excited alkali atoms². A typical optical spectrum taken at room temperature is shown in Fig. 3. Here, a 300 eV electron beam was incident along the surface normal on a single crystal LiF sample which had been cleaved in air and baked under UHV conditions at 600°C. Fluorescence radiation was collected at 90° from the surface normal; the ambient pressure was less than 10^{-9} Torr. The 6708 Å lithium resonance line is clearly visible as well as a trace signal of hydrogen Balmer alpha at 6563 Å. As in the photon irradiation studies, no other excited lithium lines were observed above the background noise; any would have to be at least a factor of 100 weaker than the observed line.

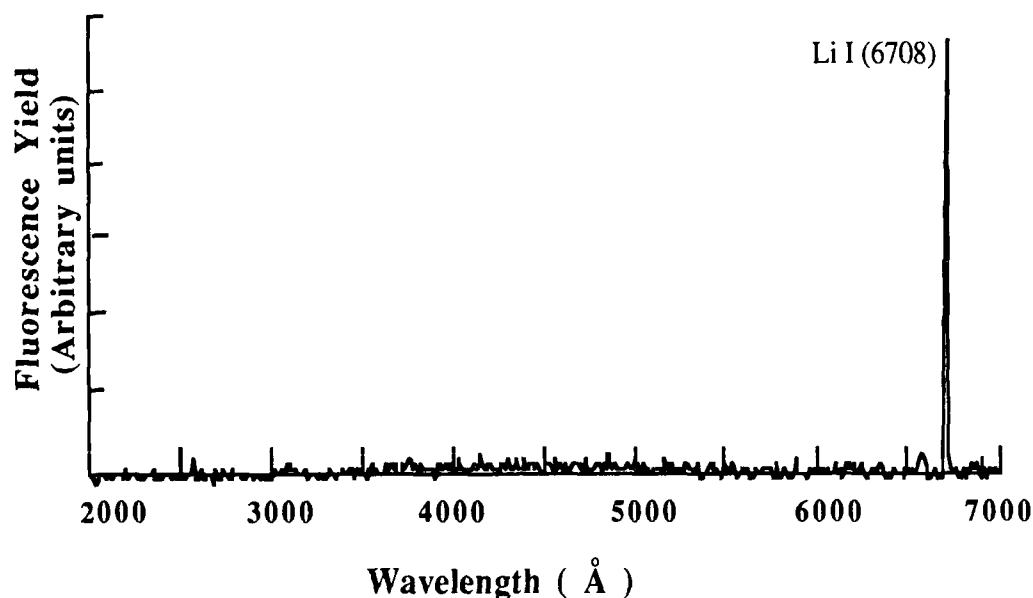


Fig 3. Fluorescence spectrum of LiF bombarded with 300 eV electrons

D. Lithium Metal Studies

We have observed energetic electron and photon induced desorption of excited lithium neutrals from a thickly dosed lithium film, which was possibly contained oxygen. The experimental setup was similar to that described above. In vacuum we dosed a soda-lime glass plate by means of a SAES lithium getter. Since the dosed film was opaque (it appeared shiny gray), we conclude from the attenuation length of light in lithium that it had a thickness of at least 1000 Å. At a pressure of 10^{-9} Torr we observed electron stimulated desorption of Li^* (6708 Å) within 5 minutes of deposition. The electron beam current was 80 μA and energy 300 eV. Figure 4 shows a fluorescence spectrum taken with the sample at room temperature. The spectrum represents 200 signal integrations over five minutes at 1 mm slit width. Previously observed bulk fluorescence of the glass plate had vanished; the plate is viewed as a mechanical support only. The power density of the electron beam was 10 mW/mm^2 which does not cause appreciable heating or thermal evaporation of the lithium layer.

In another study, we used a tungsten substrate. The base pressure was 10^{-11} Torr, thus the environment was much "cleaner" than that for the study described above. The lithium resonance

line was not seen until several hours after the dosing. Subsequently, zero order synchrotron radiation was used to desorb Li^* . The results show exactly the same characteristics as those for electrons.

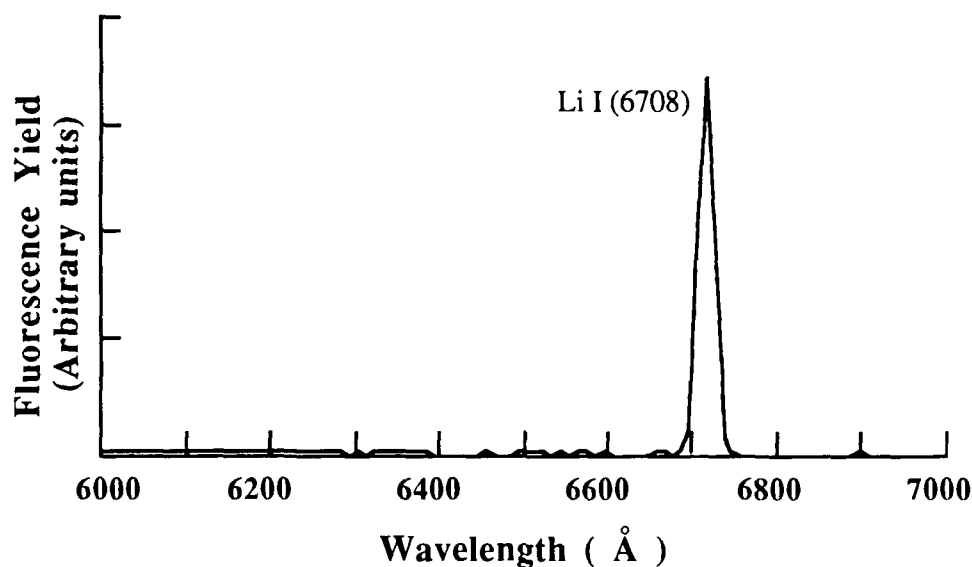


Fig 4. ESD of Li^* from a lithium layer dosed on glass

E. Discussion

In the DIET experiments (Figs. 1, 3 and 4), we observe only the first lithium resonance line. These results contrast with the ion sputtering results of Fig. 2, where emissions from higher excited states are observed. We therefore propose the following picture.

It has been well established that the surface does not remain stoichiometric during electron or photon induced desorption of alkali halides at room temperature. The halide atoms desorb much faster leaving behind an alkali enriched surface³. In our ESD and PSD studies of LiF , we thus expect to have excited lithium atoms desorbing from a surface enriched with lithium metal by a previously discussed mechanism⁴. In the desorption studies of dosed lithium, excited lithium is also desorbing in the presence of a metallic lithium surface. In this case, contamination provides centers of localization for the electronic energy deposited by the incoming electrons or photons leading to desorption of lithium.

We now account for the fact that only the 2p-2s transition is observed in the DIET experiments: as Li^* leaves the surface it interacts with the band structure of lithium metal surrounding the desorption site (Fig. 5). Electrons in the 2p level cannot resonantly tunnel to the filled levels of the metal below the Fermi energy. Excitations of Li^* to the $n \geq 3$ levels may resonantly ionize (10^{-15}s) faster than de-excitation (10^{-9}s). Hence, higher transitions are suppressed and we observe only the single 2p-2s line in our spectra.

In contrast to this, we do see higher excited states of lithium upon ion bombardment of LiF. Ion sputtering is much more violent than desorption induced by electronic transitions and proceeds through momentum exchange processes. Under ion bombardment, the surface is more microscopically rugose and a lithium metal band structure will be much less developed. The desorbing Li^* departs with a higher velocity and resonant de-excitation of higher excited states by interaction with the band structure is less probable. Consequently, we are able to observe radiative transitions from the higher excited states of lithium.

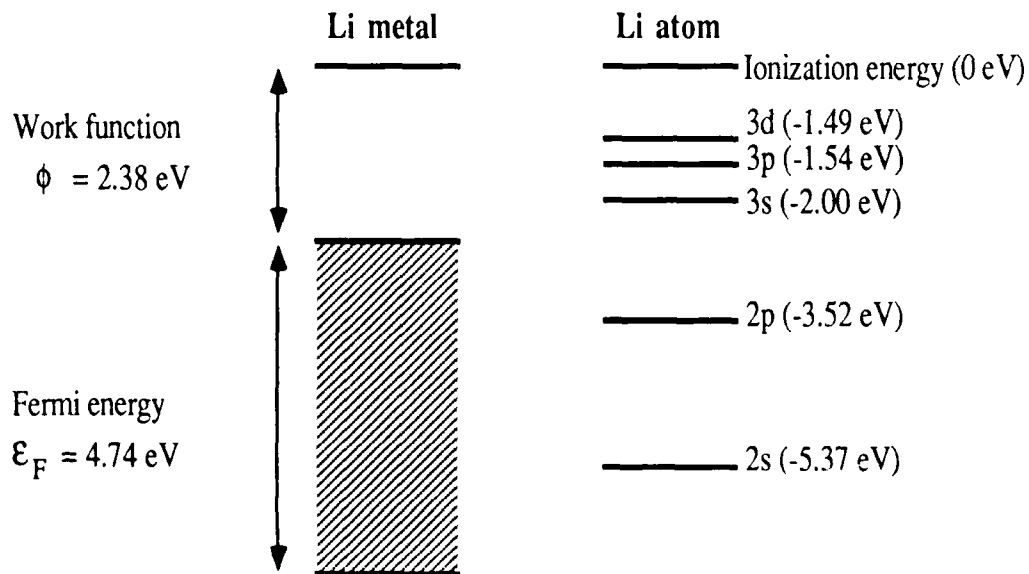


Fig. 5. Schematic energy level diagram of a lithium atom near lithium metal.

F. Conclusions

On the basis of the work performed under this grant, the following conclusions may be made:

(A). It is clear that desorption induced by electrons and photons proceeds by different mechanisms than does desorption induced by ion bombardment. For the ESD and PSD processes occurring at LiF and lithium-dosed surfaces at room temperature, the presence of a metal rich surface provides a channel for de-excitation of excited lithium states which are above the lithium metal Fermi energy. This quenching of the higher excited lithium lines is less likely when the sample is excited by ions⁵.

(B). This research has dramatically pointed out the importance of thin surface layers on the final states of desorbed species. Consequently, theoretical^{6,7} and experimental⁸⁻¹⁰ work is presently under way to assess the influence of hydrogen and other overlayers on these processes.

(C). The importance of this approach to further understanding of surface reactions in space and damage in optical systems can not be overstated¹¹. Clearly, this is an instance where fundamental studies have significant impact on recognized national needs.

Further studies will include careful surface analysis of the stoichiometry of lithium fluoride, time dependence, and beam energy dependence during ion, electron and photon irradiation. The ultimate aim of these studies is to characterize the final states of all the desorption products and the extent to which they are influenced by the surface, exciting beam and secondary processes. AFOSR is currently supporting work in these research areas through research contracts.

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9. M. Mendenhall, A. Barnes, P. Bunton, R. Haglund, L. Hudson, R. Rosenberg, D. Russell, J. Sarnthein, P. Savundararaj, N. Tolk, and J. Tellinghuisen, *Chem. Phys. Letters* 147, 59 (1988)
10. L. T. Hudson, A. V. Barnes, N. J. Halas, R. F. Haglund, M. H. Mendenhall, P. Nordlander, N. H. Tolk, Y. Wang, and R. A. Rosenberg, in *Desorption Induced by Electronic Transitions, DIET 3*, ed. by R. Gomer (Springer-Verlag, Berlin, 1988) p. 274
11. R. F. Haglund, Jr., *SPIE* 895, 182 (1988)

3. Personnel

Senior personnel employed under this project included, in addition to the PI's: Professors R. Albridge, J. Tellinghuisen, and A. Barnes; and Drs. D. Russel and P. Nordlander. Junior personnel included the following graduate student Research Assistants: D. Harper, P. Savandararaj, M. Shea, and S. Oyog. In addition B. Pruit, W. Zicko, C. Ricafort, T. Peatman, E. Moore, N. Westlake, K. Haglund, V. Salota, D. Kaplan, and P. Bhorjee were employed at various times as undergraduate research assistants. A collaborative interaction has evolved with personnel from the Institute for General Physics, Technical University, Vienna, Austria. The researchers from Vienna are Drs. Wolfgang Husinsky and Gerhard Betz. A strong theoretical collaboration continues to exist with Dr. John C. Tully of AT&T Bell Laboratories, Murray Hill, New Jersey.

4. Consultative and Advisory Functions (Norman Tolk)

- Consultant to Institute for Defense Analysis, 1988-
- Consultant to Chemistry Division, Los Alamos National Laboratory, 1986-
- Consultant to S-Cubed Corporation
- Consultant to Lockheed Corporation, 1987

- Consultant to Acurex Corporation, 1986-
- Consultant to Eaton Corporation, 1987-

5. Publications

"Erosion and Glow in the Near-Earth Space Environment," R. G. Albridge, R. K. Cole, A. F. Daech, R. F. Haglund, Jr., C. L. Johnson, H. Pois, P. M. Savundararaj, N. H. Tolk, and J. Ye, Nuclear Instruments and Methods in Physics Research B18, 582 (1987)

"Threshold Effects and Time Dependence in Electron- and Photon-Stimulated Desorption," Richard F. Haglund, Jr., Norman H. Tolk, G. M. Loubriel, and Richard A. Rosenberg, Nuclear Instruments and Methods in Physics Research B18, 549 (1987)

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"Optical Phenomena Observed in Low-Energy Ion and Electron Bombardment of Silica Surfaces," H. C. Mogul, R. F. Haglund, R. A. Weeks, D. L. Kinser, N. H. Tolk, and P. Wang, Submitted to Journal of Vacuum Science and Technology A (December 1987)

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"Intracavity Optical Damage Due to Electrons, Ions and Ultraviolet Photons," R. F. Haglund, Jr., *Proceedings of the Society of Photo-Optical Instrumentation Engineers* 895, 182 (1988)

"Extended Huckel Theory for Ionic Molecules and Solids: An Application to Alkali Halides," Y. Wang, P. Nordlander, and N. H. Tolk, to be published in the October 1988 issue of *J. Chem. Phys.*

"Competition Between Bulk and Surface Excitations in Photon-Stimulated Desorption of Li^* from LiF ," R. F. Haglund, Jr., A. V. Barnes, M. H. Mendenhall, N. H. Tolk, L. Hudson, and D. A. Ramaker, to be submitted to *Physical Review Letters* (1988)

6. Papers Presented at Meetings, Conferences, Seminars, Etc.

"Dynamical Responses to Light in Crystalline and Polymeric Materials," R. F. Haglund, Jr., Abstract submitted to the ONR FEL Contractors Meeting (May 1987)

"Energy Shifts and Broadening of Excited H Levels in the Vicinity of a Metal Surface," P. Nordlander and J. C. Tully, AT&T Bell Laboratories Abstract (1988)

"Anomalies in the Velocity Distributions of Excited Metal Atoms Sputtered from Metals and Their Oxides," G. Betz, P. Wurz, W. Husinsky, H. Stori, B. Strehl, E. Wolfrum, N. H. Tolk, P. Nordlander, and R. F. Haglund, Jr., Abstract (1988)

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"Coherence, Orientation and Alignment Processes in Charge Transfer at Solid Surfaces" N. H. Tolk, Invited Talk, March Meeting of the American Physical Society, 4/3/86

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