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**Electronic Interactions of Directed Photon Beams  
with Surfaces**

contract: F49620-88-C-0080

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Final Report for the period June 1, 1988 - August 31, 1990

Prepared for

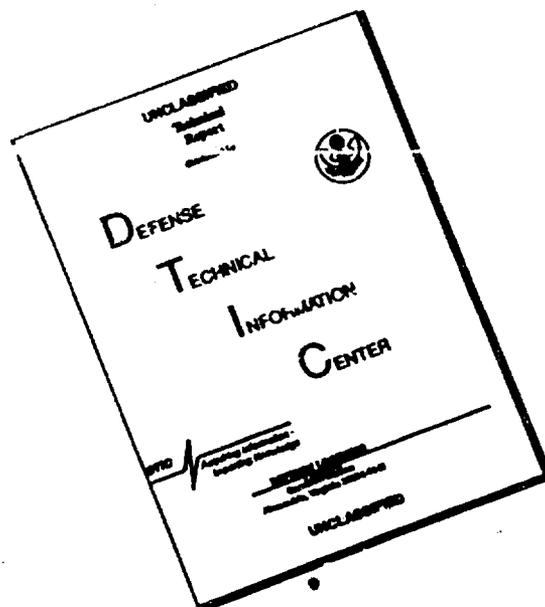
*Air Force Office of Scientific Research  
Directorate of Chemical and Atmospheric Sciences  
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91-13679

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# REPORT DOCUMENTATION PAGE

QMB No. 0704-0128

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1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE September 1990		3. REPORT TYPE AND DATES COVERED Comprehensive Final Report June 1, 1988 to August 31, 1990	
4. TITLE AND SUBTITLE Electronic Interactions of Directed Photon Beams with Surfaces				5. FUNDING NUMBERS	
6. AUTHOR(S) Alan V. Barnes, Marcus Mendenhall and Norman H. Tolk					
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Department of Physics and Astronomy Center for Atomic and Molecular Physics at Surfaces Vanderbilt University Box 1807, Station B Nashville, TN 37235-1807				8. PERFORMING ORGANIZATION REPORT NUMBER 4-20-630-4183	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Lt. Col. Larry Burggraf, Program Manager Directorate of Chemical and Atmospheric Sciences Air Force Office of Scientific Research Department of the Air Force Bolling Air Force Base, DC 20332-6448				10. SPONSORING/MONITORING AGENCY REPORT NUMBER F49620-88-C-0080	
11. SUPPLEMENTARY NOTES					
12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release Distribution unlimited				12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words)  The object of this research program is to investigate the fundamental dynamics of energy-surface interactions, with emphasis on the ways in which the energy deposited by incident uv photons and electrons lead to surface bond-breaking phenomena, to fluorescence, and to modification of surfaces and near-surface layers of bulk materials. Optical radiation may arise from electronic excitations in the near surface bulk or from excited atoms and molecules emitted from the surface. Our studies of electronically- induced desorption have shown that a significant fraction of the incident energy may be channeled into bond-breaking and desorption processes leading to ejection of excited neutral atoms and molecules from metal oxide and insulator surfaces which subsequently decay to produce optical radiation in the visible, UV or IR. Here, we report on (a) photon-stimulated desorption of excited alkali atoms following irradiation of alkali halide crystals with ultraviolet light and on (b) optical fluorescence arising from MgF <sub>2</sub> , Be, Al <sub>2</sub> O <sub>3</sub> , and BeO surfaces using an electron beam at room- and at cryo- temperatures under different gas dosing environments.					
14. SUBJECT TERMS				15. NUMBER OF PAGES 15	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED				16. PRICE CODE	
18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED		19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED		20. LIMITATION OF ABSTRACT SAR	

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## 1. Research Objectives

The object of this research program is to investigate the fundamental dynamics of electromagnetic beam interactions with surfaces, with emphasis on the ways in which the energy deposited by incident uv photons lead to surface bond-breaking phenomena and to modification of surfaces and near-surface layers of bulk materials. When energetic photons or electrons impact on surfaces, many complex interrelated processes occur as the incident energy is transformed into fluorescence and other electronic excitations. Optical radiation may arise from electronic excitations in the near surface bulk or from excited atoms and molecules emitted from the surface. Recent studies of electronically- induced desorption have shown that a significant fraction of the incident energy may be channeled into bond-breaking and desorption processes leading to ejection of excited neutral atoms and molecules from metal oxide and insulator surfaces which subsequently decay to produce optical radiation in the visible, UV or IR. On the ionic insulators studied, erosion of excited neutral atoms and molecules mediated by the migration of electronically created defects into the surface and near-surface layers is the dominant phenomenon, and in some cases occurs with nearly unit efficiency.

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. We report photon-stimulated desorption of excited alkali atoms following irradiation of alkali halide crystals with ultraviolet light of valence-band energies and the radiation damage on  $MgF_2$ , Be mirror,  $Al_2O_3$ , and BeO surfaces using an electron beam at room- and at cryo- temperatures under different gas environments.

## 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. Bending magnet radiation from an 800 MeV electron storage ring was dispersed either by a Seya-Namioka (SN) Rowland-type monochromator or by a toroidal-grating monochromator (TGM) and focused onto a sample at normal incidence in an ultra-high vacuum chamber with a base pressure generally around  $10^{-10}$  Torr. Useful photon flux from the monochromator was obtainable over the energy range 8-28 eV. 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 2000-8000 Å using a 1200 lines/mm grating blazed at 5000 Å. Fluorescence radiation was measured at 90° to the surface normal. For each point in the excitation function, the signal was counted at the peak of the first resonance line of the desorbing excited Li atoms ("on resonance"), and then counted at wavelengths "off resonance" on both sides of the atomic emission line for background subtraction. For each run, the storage ring current and the current recorded on a nickel mesh positioned to monitor the flux reflected from the grating were recorded for normalization.

Samples used in this experiment were alkali halide single crystals, most obtained from Harshaw Chemical Company; some of the measurements on LiF were made on samples from Optovac. The samples were mounted on a copper target holder attached to a Huntington micromanipulator, and were usually under vacuum within about an hour after being cleaved in air. Following installation of the samples in the UHV chamber, the vacuum system was then baked at 200°C for several hours, and the samples were subsequently cleaned by heating a few hours at 300°C to 400°C.

Figure 1 shows two fluorescence spectra observed during uv-photon irradiation of LiF at photon energies of 13.5 eV and 21.5 eV in the Seya monochromator. The first resonance line (671nm) of neutral lithium is clearly evident. This is clear evidence that the desorption is produced by valence-band excitation. An excitation function of desorbed excited lithium from LiF is shown in figure 2(a). The data have been normalized to the Ni mesh current and to the response of Ni. The curve plotted through the experimental points was obtained by smoothing the data. The uv reflectance spectrum and absorption data show similar structure for photons in this energy range.

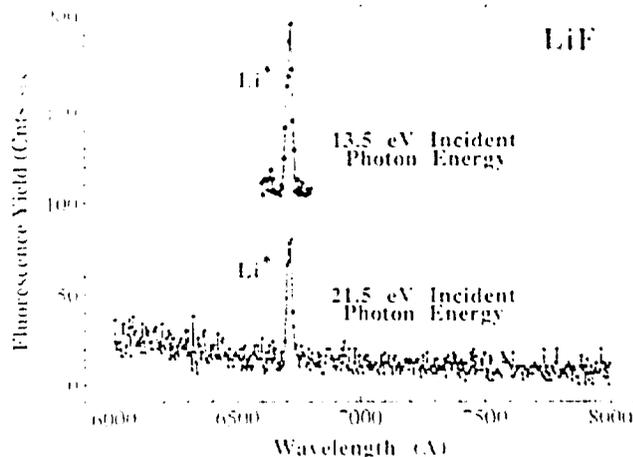
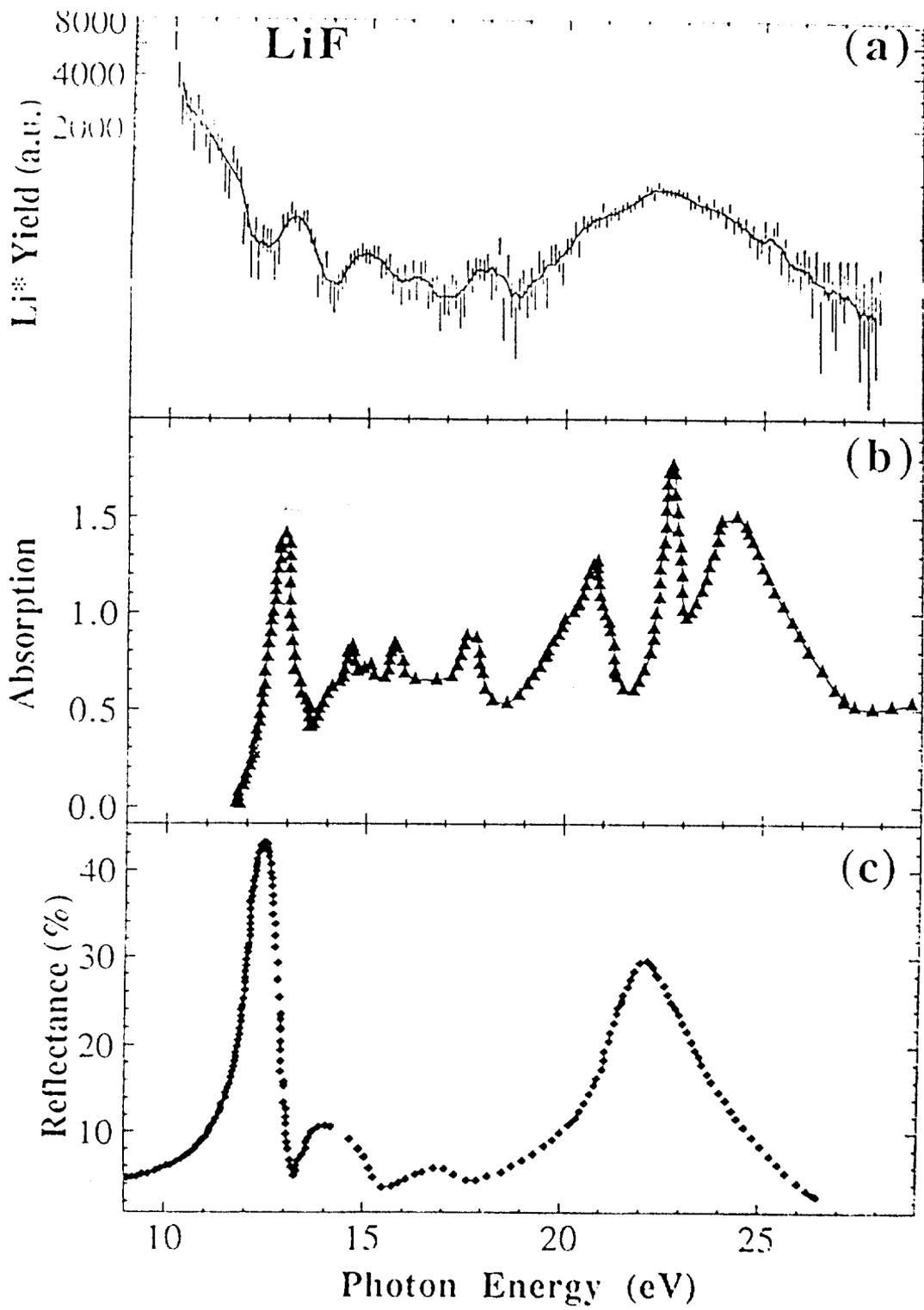


Figure 1. Fluorescence scan from PSD of LiF at 13.5 eV and 22.5 eV, showing the optical signal from the 2p to 2s transition of the Li atom.



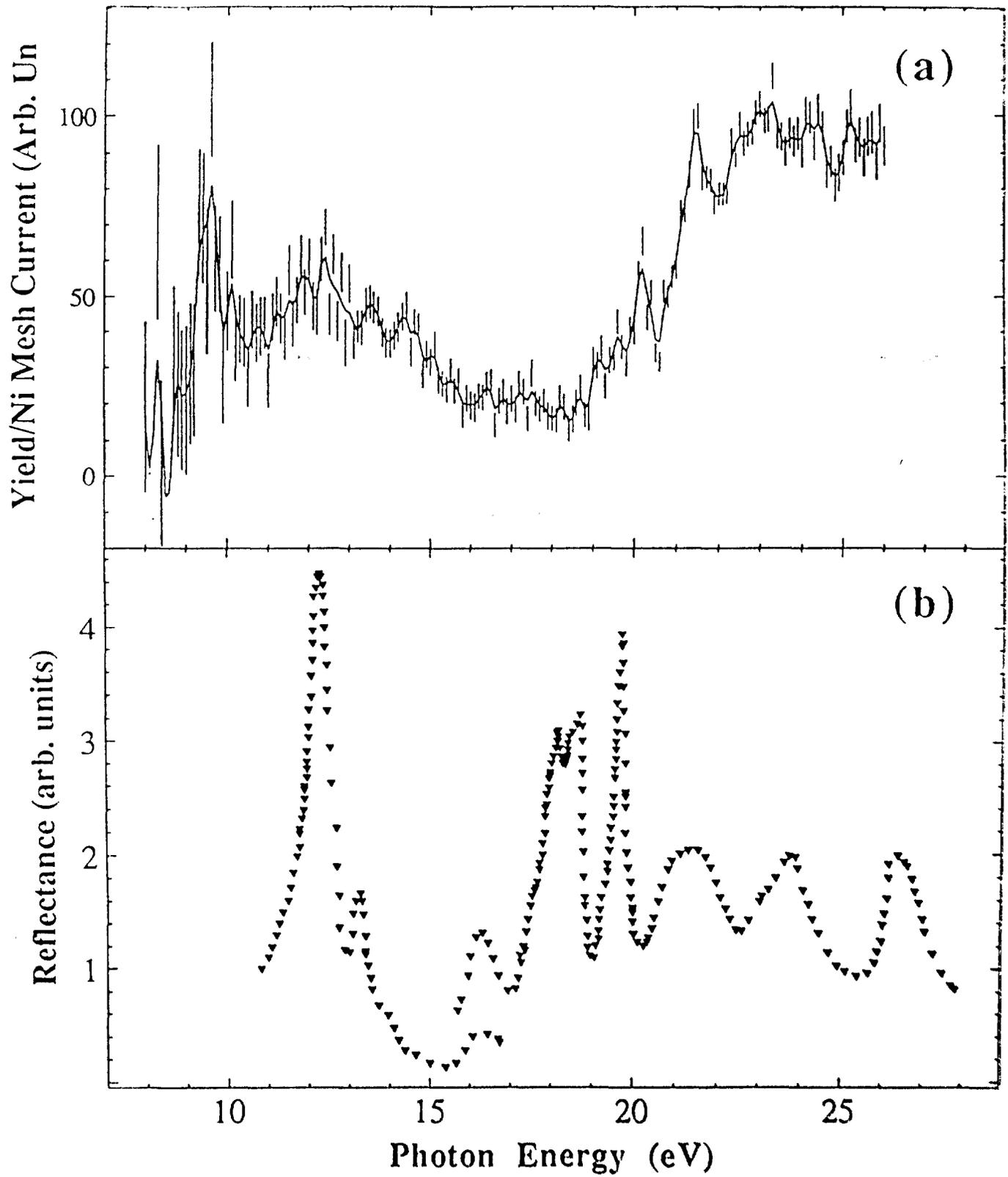
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Figure 2. (a) Excitation function of Li\* from LiF over the incident photon energy range 8-28 eV. (b) Absorption spectrum of LiF measured in a single crystal, digitized from original data in Reference 1. (c) Reflectance spectrum from a thin film of LiF, digitized from Reference 2.

B. PSD of  $K^*$  from Potassium Chloride

Excited potassium atom desorption was studied under the same conditions as for LiF. Figure 3(a) is an excitation for the production of excited potassium under photon irradiation of KCl.



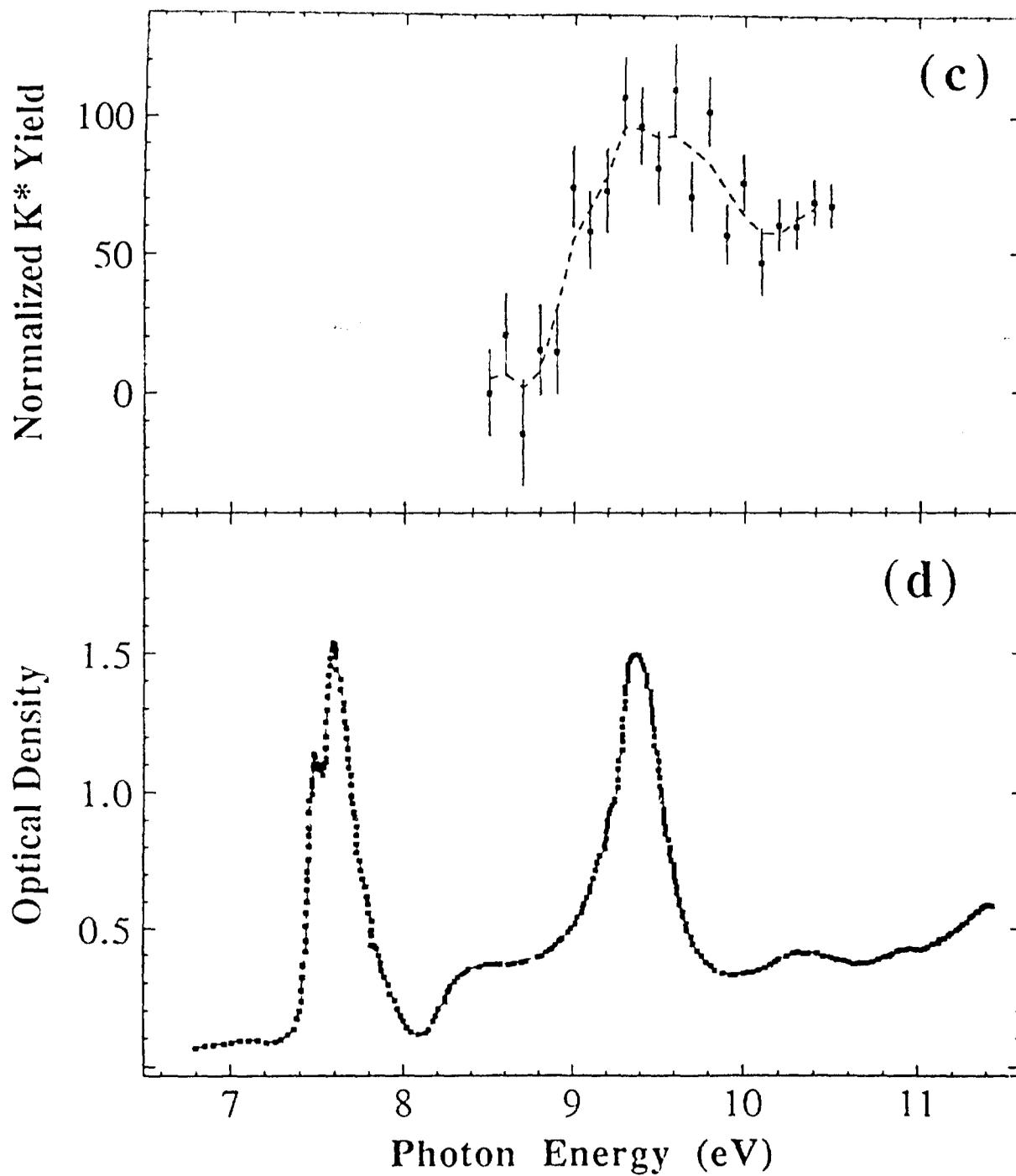


Figure 3. (a) Excitation function of  $K^*$  (767 nm 4p to 4s transition) from KCl. (b) Absorption data from KCl, from Reference 3. (c) Excitation spectrum of  $K^*$  from KCl for photons near the

surface exciton peak. (d) Reflectance spectrum from KCl near the surface exciton peak, adapted from reference 4.

### C. Glow Data in the UV and Visible Regions

Figure 4 shows the glow stimulated by 200 eV electrons, from three samples, Al<sub>2</sub>O<sub>3</sub>, MgF<sub>2</sub> and Be, at 65 K without any gas dosing in the ultra high vacuum chamber. The pressure in the UHV chamber with operating electron gun was  $2 \times 10^{-9}$  Torr. The typical fluorescence spectrum consists of several sharp lines sitting on a broad background. These sharp lines are the light emission due to the deexcitation processes of the desorbed species. The most intense line at 3883 Å was identified to be CN neutral molecule which appears on all three samples. There are some further small lines, i.e. OH\* at 3080 Å, H<sub>α</sub> at 6563 Å and other CN vibrational bands. The broad band below these sharp lines is the electron stimulated fluorescence originating from the bulk. Since the 200 eV electrons penetrate only a few hundred angstroms, this fluorescence band occurs only in the near surface part of the crystal. Besides the similarities of desorbing species each substrate shows its own characteristic features concerning the yield and relative contribution of the bulk glow. The biggest fluorescence yield from desorbing atoms and molecules is observed on the Al<sub>2</sub>O<sub>3</sub> sample, whereas the bulk glow on MgF<sub>2</sub> is much more intense than on the other two samples.

### D. Dosing dependence

Different gases, such as N<sub>2</sub>, H<sub>2</sub>, O<sub>2</sub> or air, have been introduced into the UHV chamber at a base pressure of  $2.0 \times 10^{-9}$  Torr. The spectra were measured at a constant partial pressure of  $1.0 \times 10^{-7}$  Torr, constant sample temperature of 65 K, and a constant electron energy of 200 eV.

The gas dosing effect on Al<sub>2</sub>O<sub>3</sub> and BeO are shown in Figure 5 and Figure 6 respectively. In the case of N<sub>2</sub> dosing, the desorption yield of CN\* is considerably increased, at least on the Al<sub>2</sub>O<sub>3</sub> sample. By dosing hydrogen, the Balmer lines H<sub>α</sub> and H<sub>β</sub> appear rather strongly and also the OH\* signal is enhanced. Introducing air into the UHV chamber leads to a general increase of all signals. The spectra in Figure 5 and Figure 6 clearly show, that the glow and the desorption yield are substantially increased due to cryolayer coverage.

### E. Temperature dependence

Glow and desorption was detected at different sample temperatures by using the same electron energy and same dosing gases. Since the electron current was not normalized, only the

general trend can be described here. The general trend is that the glow intensified by reducing the sample temperature. The desorption yield is also enhanced by reducing the sample temperature.

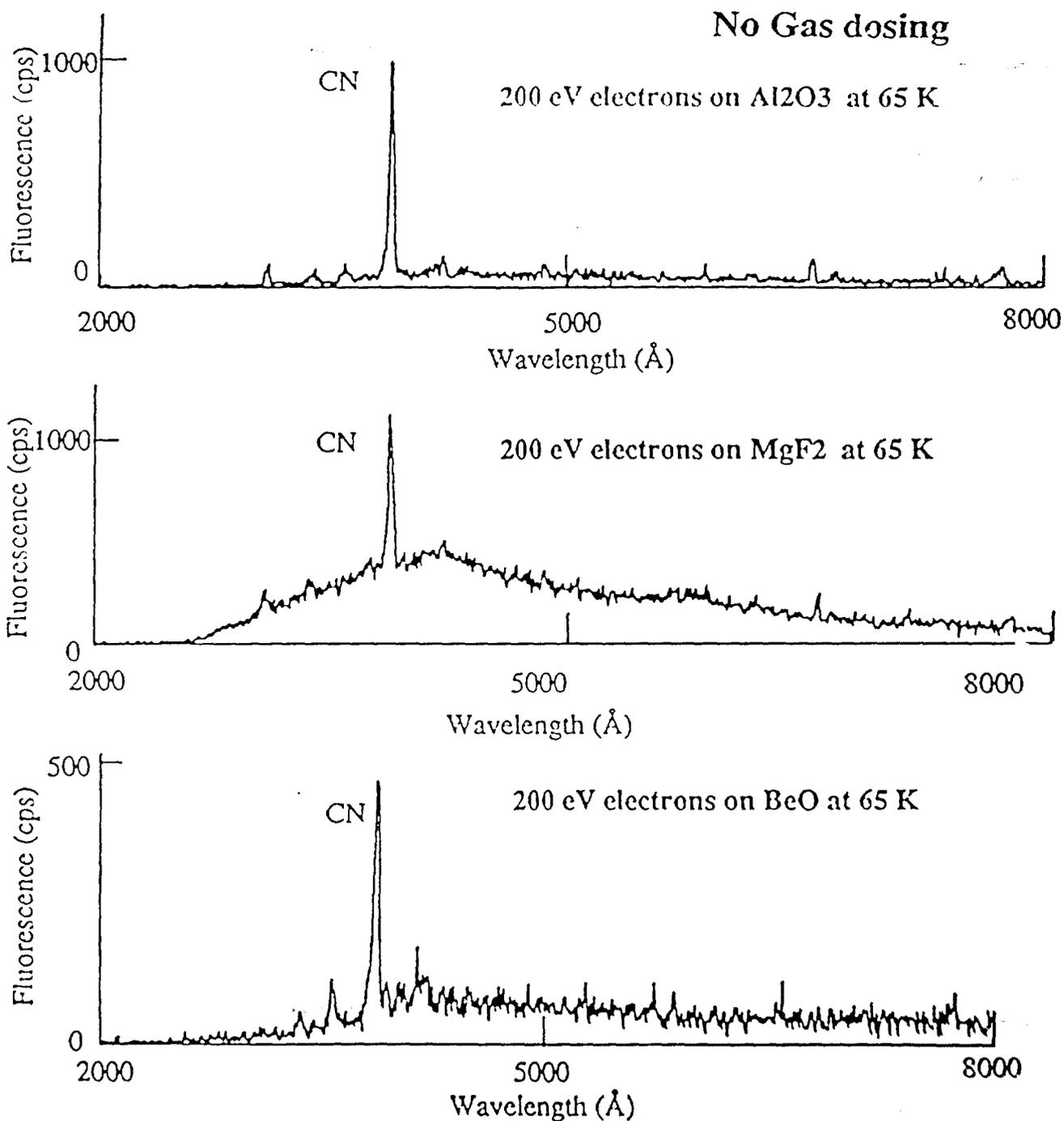


Figure 4 Fluorescence induced by 200 eV electrons impacting on Al<sub>2</sub>O<sub>3</sub>, MgF<sub>2</sub> and BeO surfaces at 65 K

### 200 eV electron on Al<sub>2</sub>O<sub>3</sub> at 65 K

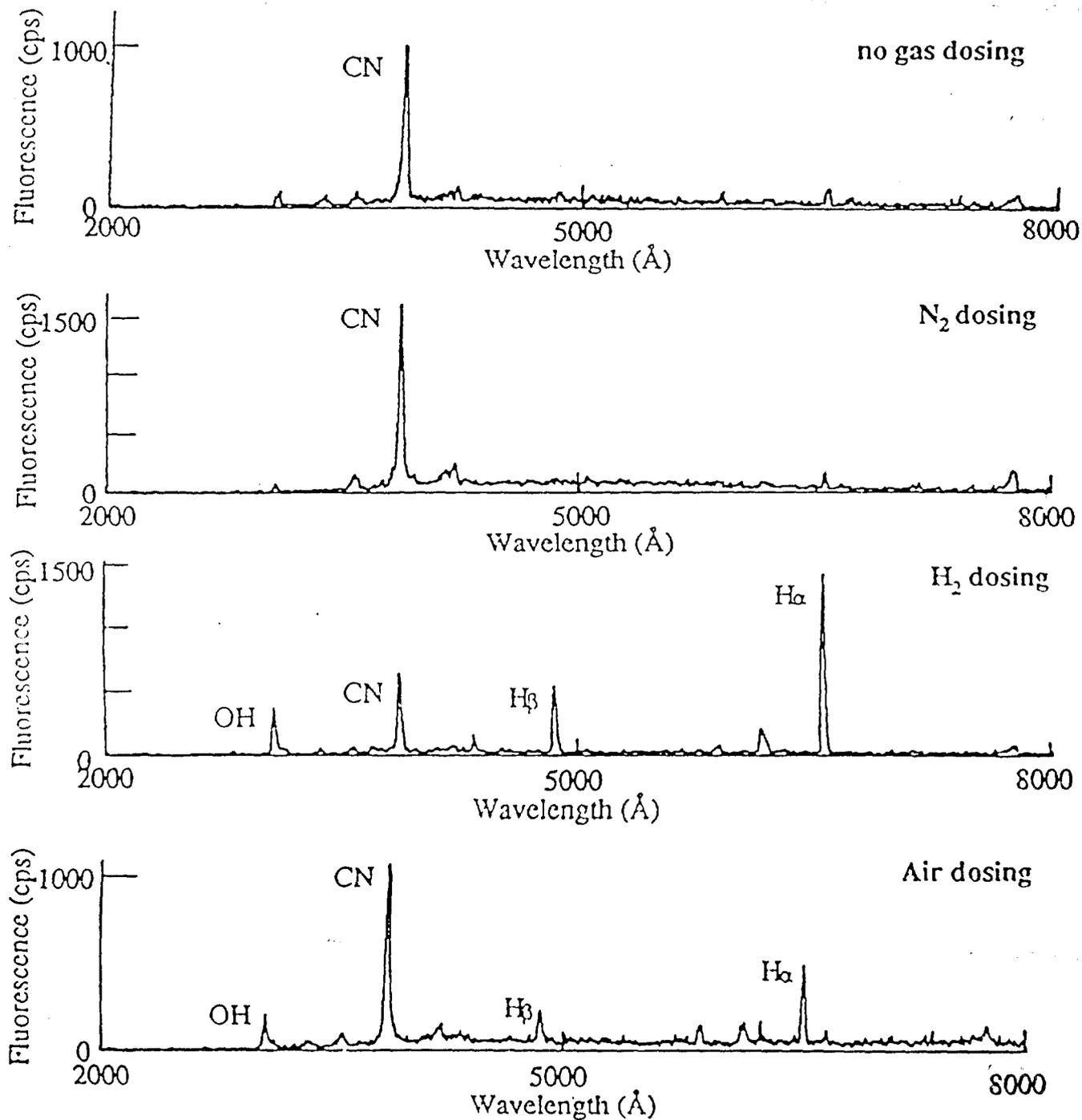


Figure 5 : Dosing effect on the glow from a Al<sub>2</sub>O<sub>3</sub> surface

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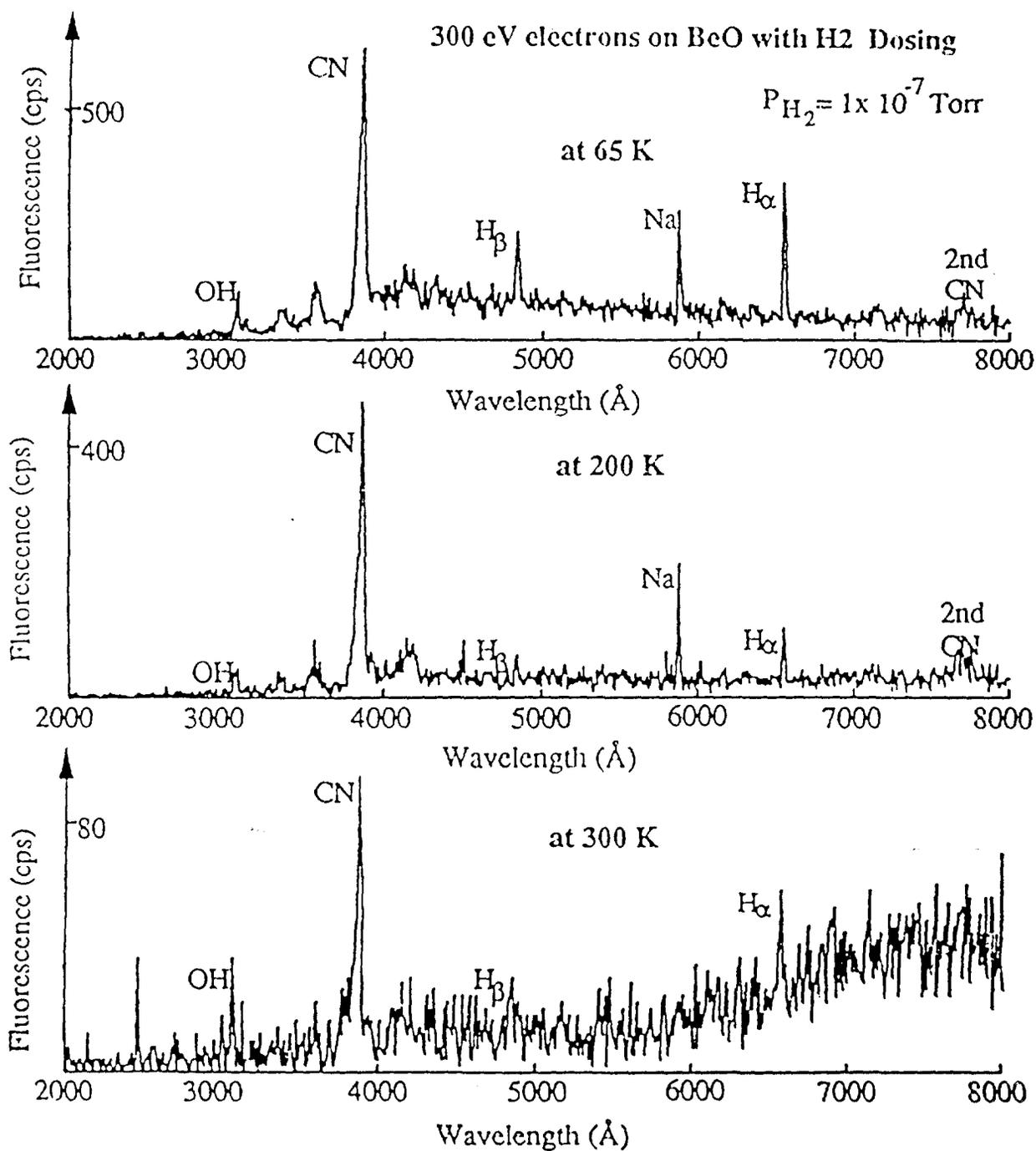


Figure 6: Glow induced by 200 eV electrons on BeO at 65 K, 200 K and 300 K

#### F. Energy dependence

The energy dependence of the fluorescence yield is most clearly shown on the  $\text{MgF}_2$  sample. The glow was monitored from  $\text{MgF}_2$  by changing the energy of the incident electrons at the same experimental parameters except the energies of the electrons. The glow is gradually increased with increasing electron energy.

#### G. Discussion

The foregoing  $\text{Li}^*$  and  $\text{K}^*$  experimental results are not consistent with the most widely invoked mechanisms for PSD of excited atoms. One model involves ion neutralization, and is analogous to the Knotek-Feibelman mechanism for ion desorption. According to this mechanism, the interatomic Auger decay of a core hole results in the formation of two valence holes on a single surface anion resulting in ion desorption<sup>5,6</sup>. Subsequent neutralization of the desorbing ion into an excited state then accounts for the observed excited atom yield. The second mechanism<sup>7</sup> assumes that the excited atoms are produced by secondary-electron excitation of desorbed ground state atoms in the gas phase. Our experiments show exclusively that excited-atom desorption can be initiated by a one-photon transition from the valence band. The excitonic features in the PSD yields do not rule out the production of excited atoms by secondary-electron excitation of desorbed ground state alkali atoms. However, the measured temperature dependence clearly demonstrates that secondary electron excitation of desorbed ground state atoms is not the source of the present observations, since the yields of ground-state alkali atoms generally increase with temperature above the temperature for vaporization of the alkali metal, while the excited atom yields measured in these experiments decreases with temperature.

#### H. Conclusions

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

(A). We have described the first observation of photon-stimulated desorption of excited alkali atoms from alkali halide crystal surfaces for photons with valence-band energies. The excitation functions of excited-atom yields are correlated with excitonic structure observed in previous reflectance and absorption measurements for both valence-band and core-level excitation<sup>8</sup>.

(B). This research has dramatically pointed out the importance of thin surface layers (by gas dosing) on the final states of desorbed species and surface glow. Consequently, theoretical<sup>9,10</sup>

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and experimental<sup>11-16</sup> work is presently under way to assess the influence of hydrogen, nitrogen, 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 cannot be overstated<sup>14</sup>. Clearly, this is an instance where fundamental studies have significant impact on recognized national needs.

Further studies will include characterization of the final states of all the desorption products and the extent to which they are influenced by the surface and surface layers, beam energy and substrate temperature.

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### 3. Personnel

Senior personnel employed under this project included, in addition to the PI's: Professor R. Albridge, Dr. D. Russell, Dr. P. Nordlander Dr. Manfred Riehl-Chudoba, Dr. J. Binet. Junior personnel included the following graduate student Research Assistants: L. T. Hudson, P. Bunton, J. Sarnthein, P. M. Savundararaj, and D. Liu. In addition D. McClure, and M. Albert, were employed 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)

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

## 5. Publications

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

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"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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"Electronic Interactions at Surfaces," N. H. Tolk, Chemistry Seminar, Yale University, 4/23/87

"Interactions of Photons, Electrons and Heavy Particles with Surfaces," N. H. Tolk, Physics Colloquium, University of Arkansas, 4/24/87

"Electronic Desorption Processes," N. H. Tolk, Invited Talk, International Workshop on Desorption Induced by Electronic Transitions, DIET-III, Long Island, New York, 4/21/87

"Electronic Processes at Surfaces," N. H. Tolk, Physics Colloquium, Georgia Institute of Technology, 10/28/87

"Mechanisms of Energy-Surface Interactions," N. H. Tolk, Physics Colloquium, Institute for Defense Analysis, Washington, D.C., 1/25/88

"Electronic Desorption Processes," N. H. Tolk, Invited Talk, 4th International Symposium on Resonance Ionization Spectroscopy, N.B.S., Gaithersburg, MA, 4/20/88

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