Project Title: Scattering of Hydrogen From Surfaces

Project No: G-41-655 (Follow-on to G-41-648)

Project Director: Dr. E. W. Thomas

Sponsor: Energy Research & Development Admin.; Oak Ridge Operations; Oak Ridge, TN 37830

Agreement Period: From 3/1/77 Until 2/28/78 (Contr. Period)

Type Agreement: Contract No. EY-76-S-05-2591; Mod. No. A019 (Formerly E(40-1)-2591)

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Contractual Matters
(thru OCA)

Defense Priority Rating: None

Assigned to: Physics (School/Laboratory)

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Other
Project Title: Scattering of Hydrogen From Surfaces

Project No: G-41-655

Project Director: Dr. E.W. Thomas

Sponsor: U.S. Dept. of Energy; Oak Ridge Operations, P.O. Box E; Oak Ridge, TN 37830

Effective Termination Date: 2/28/78 (Mod. No. A019 Period)

Clearance of Accounting Charges: by 2/28/78

Grant/Contract Closeout Actions Remaining:

- Final Invoice and Closing Documents
- Final Fiscal Report (Cert. Stmt. of Costs per Appendix "C") due by 5/31/78
- Final Report of Inventions
- Govt. Property Inventory & Related Certificate
- Classified Material Certificate
- Other

Follow-on Project is G-41-672.

Assigned to: Physics (School/Laboratory)

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Project File (OCA)
Project Code (GTRI)
Other
SCATTERING OF HYDROGEN FROM SURFACES

PROGRESS REPORT NO. 1

Covering the Period

November 1, 1976 to November 1, 1977

by

E. W. Thomas

E. O. Rausch

J. E. Harriss

W. A. Metz

A. J. Senol

U. S. Energy Research and Development Administration

Report No. ORO-2591-81

Contract No. AT-(40-1)-2591

U. S. Energy Research and Development Administration

Oak Ridge, Tennessee

1 November 1977
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I. Title

Scattering of Hydrogen from Surfaces

II. Contract Number

This report summarizes work performed on excitation phenomena under contract AT-(40-1)-2591 for the U. S. Energy Research and Development Administration. The present report covers the period 1 November 1976 to 1 November 1977 which corresponds to the first 8 months of the 12 month period covered by modification No. 19 to this contract, plus the final four months of the preceding contract period.

IV. Abstract

Scattering of hydrogen ions and atoms from surfaces is the principal subject studied, with some limited supporting research on sputtering, surface chemical reactions, and on the scattering of non-hydrogenic particles.

Our previous studies of excited hydrogen backscattering from metals have been extended to cover an impact energy range from threshold (about 1 keV) to 200 keV and to include incident molecular ions. We have a complete model of the behavior which includes Auger de-excitation processes and also an electron ion recombination mechanism to explain neutral atom formation. The model has been shown to explain changes to neutral atom formation when oxygen is adsorbed on metal surfaces and the model has also been tested with somewhat heavier projectiles (B⁺).

We have evaluated the potential value of the SCANIIR technique (Surface Composition Analysis by Neutral and Ion Induced Radiation) for in-situ analysis of the first wall in a plasma device and concluded that it is not yet suitable for detection of contaminants such as H, C and O.
Direct measurements of energy spectra for recoil $H^+$ emerging at small angles to the surface show peak fluxes at 8 keV recoil energy which differs from the 1 to 2 keV maximum reported previously for large angle scattering. Fluxes of $H^+$ and $H^-$ have also been measured directly.

IV. Discussion of Progress

A. Introduction

The topics from the "Proposed Technical Program" of our previous proposal were as follows.

Project (a) Measurement of the flux and charge state composition of particles backscattered when $D^+$, $H^+$, $H_2^+$ and $H_3^+$ are incident with 1 to 30 keV/nucleon energy on Be, Cu, Mo and Stainless Steel; assessment of changes to charge state distributions when adsorbed $O_2$ is present. Target date 29 February 1978. Percentage of contract time devoted to project 50%.

Project (b) Study of excited H backscattering when 20 to 200 keV $H^+$ and $D^+$ are incident on Cu, Mo and Stainless Steel to test theoretical predictions. Target date 29 February 1978. Percentage of contract time devoted to project 20%.

Project (c) Assessment of SCANIIR technique for detection of H, C, O and metals, present as contaminants in Be, Mo, Au and Stainless Steel; determination of conditions for optimum sensitivity and measurement of sensitivity. Target date 30 July 1978. Percentage of contract time devoted to project 30%.
Project (a) is running a little behind schedule; many experimental difficulties have been overcome and data is now being routinely collected. We have recently increased the manpower on this project and the work with \( H^+ \) and \( D^+ \) should be largely achieved by the target date although not the work with molecular ions. Projects (b) and (c) are completed and the more significant results are in course of publication.

In addition to the proposed work we have performed the following studies.

Project (d) Investigation of how adsorbed oxygen influences the backscattering of excited \( H \) for \( H^+ \), \( H_2^+ \) and \( H_3^+ \) ions on metals.

Project (e) Investigation of whether the backscattering of excited atoms from surfaces is influenced by the temperature of that surface.

Project (f) Measurements of \( B \) and \( B^+ \) backscattering when \( B^+ \) ions are incident on selected metals.

Project (d) was a hold over from our previous contract year that required more time than we had originally envisaged; that project is now complete. We undertook the study because of evidence that the backscattering phenomena we have studied so intensively was in fact altered when surfaces were dirty; also of course all present fusion devices involve air oxidized walls with further oxide formation during oxygen discharge cleaning. It was therefore felt very important to study the effect of oxygen. Project (e) was a minor extension of our backscattering studies and followed unpublished reports from elsewhere that surface temperature influences backscattering. It was felt important to study this because the available theoretical understanding
of backscattering does not imply any significant temperature dependence; also of course any practical fusion device will involve elevated wall temperatures. The result of this brief study was that no temperature dependence could be detected. Finally project (f) was undertaken to confirm that our extensive backscattering models could be applied to ions other than H\textsuperscript{+} and He\textsuperscript{+}. The expected agreement was obtained indicating that our model’s success is not an accident related to peculiarities of the very lightest ions.

In summary, projects (b) and (c) of our original proposal and subsidiary projects (d) (e) and (f) are all complete and finished. Project (a) is running behind schedule but by concentrating our resources on this study we should achieve most of our objectives in the present contract year.

In addition to the research program we were able to substantially improve our equipment capabilities during the present year. We acquired an "ion implantation" accelerator which has extended our available energies up to 200 kV, provides beams of any element, and of course permits us to create ion implanted surfaces. Furthermore we are just assembling a sophisticated surface analysis facility that permits detailed study of surface condition during ion bombardment. This latter facility includes Auger spectroscopy and Rutherford backscattering equipment as well as capabilities for later addition of ultraviolet photoelectron spectroscopy and LEED. These new equipment capabilities represent an investment of over $150,000 and were provided through internal funds of the Institute.

A discussion follows of progress and results in these areas. For convenience the results of the subsidiary projects (d), (e), and (f) are grouped with project (b) in a general discussion of excited state formation. The narrative of the report is limited to important highlights and relevance of the results to the Magnetic Fusion Energy program; for detailed results we
shall refer to a series of Appendixes which are largely composed of published papers and articles currently under preparation.

B. Backscattering from Surfaces - Excited States

Impact of hydrogenic ions at keV energies gives rise to Doppler broadened line emission from neutral backscattered atoms. Analysis of the line shape gives information on the distributions in velocity and direction of the scattered particles. Quantitative measurements of light emission from a particular transition gives the probability of backscattered atoms being in an excited state. In the period covered by this report we have used as projectiles \( \text{H}^+ \), \( \text{H}_2^+ \), \( \text{H}_3^+ \) and \( \text{B}^+ \) at energies from 1 to 200 keV/nucleon; targets have included, Cu, Mo and Stainless Steel. Measurements were made of the \( \text{H}_\alpha (n=3+n=2) \) and \( \text{H}_\beta (n=4+n=2) \) spectral line intensities which are proportional to the flux of backscattered hydrogen atoms in the \( n=3 \) and \( n=4 \) states. In the case of \( \text{B}^+ \) impact, various spectral lines from scattered B and \( \text{B}^+ \) were monitored.

The experimental arrangement is very simple and fully described in the literature.\(^1,2\) The ion beam is produced by either a 1 to 30 kV accelerator or a 30 to 200 kV ion implanter and is directed onto a metal target at some angle \( \phi \) to the surface normal. Light emission from the point of impact is analyzed and quantitatively measured with a calibrated monochromator photomultiplier system. Since the spectral lines are Doppler broadened it is necessary to integrate over the full width of the line. Proper attention is paid to achieving atomically clean targets; generally polycrystalline metal targets are used.


In previous work with backscattered H and He we have modelled the total emission line intensity\textsuperscript{1,2,3} utilizing the following three input components:

(i) a prediction of total backscattered particle flux.

(ii) a probability that the backscattered particle is in an excited state as it recoils.

(iii) a factor to allow for radiationless de-excitation of the excited recoils by the Auger mechanism as they depart from the surface.

Our previous work was at low impact energies (1 to 30 keV) with H\textsuperscript{+} and He\textsuperscript{+} projectiles. The formulations used for the backscattering and Auger de-excitation have been discussed extensively in the literature and many de-excitation parameters evaluated\textsuperscript{1,2,3}; these aspects require no further discussion here. In this previous work we had assumed that probability of a recoil being excited was independent of recoil energy; this can be justified for low (1 to 30 keV) energies. In the present reporting period we have extended the data and the modelling to proton impact energies of 200 keV. At the higher energies (30 to 200 keV) it is necessary to allow for an energy dependent probability of the recoil being excited. This was accomplished with a theoretical prediction by Kitagawa and Ohtsuki\textsuperscript{4} which treats recoil neutralization by electron-ion recombination as the fast recoil emerges through the low electron density region at the surface. With this theoretical prediction we achieve complete agreement between experiment and theory for hydrogen impact energies from around 1 keV (the threshold below which recoils do not emerge in an excited state) to 200 keV. There is every reason to


believe that as energy is increased above 200 keV the agreement will remain excellent since the approximations made in theoretical data used for our model become more accurate. The conclusion of importance to the MFE program is that for all relevant energies (threshold to the energy of neutral beam injectors) we have a complete understanding of how excited recoils are formed at surfaces and we have confirmed a theoretical calculation\(^4\) which gives the partition of the recoil flux into ions and neutrals. The basic data are to be published shortly; the full paper is included as Appendix I and the abstract is reproduced below:


The coefficient for emission of the Balmer \(\beta\) line from backscattered H atoms has been measured for 2-200 keV hydrogen ion impact on polycrystalline targets of Mo, Cu and stainless steel. The dependence on primary energy is successfully modelled using a modification of a formulation used previously at low (2-30 keV) impact energies.

We have in the past concentrated our studies of backscattering on atomically clean metal surfaces. This of course does not necessarily represent the situation in present MFE devices where constructional materials are air oxidized and where oxygen discharge cleaning will undoubtedly produce additional oxide formation. We have therefore extended our studies to include the effect of adsorbed oxygen on the backscattering phenomenon. The general result is that the Auger de-excitation of recoils decreases, in the presence of adsorbed oxygen, permitting a greater fraction of the excited recoils to escape. From analysis of the data we can deduce coefficients for the sputtering of adsorbed oxygen from a metal surface by hydrogen ion impact; to the best of our knowledge this data has not previously been obtained by more direct techniques. The results of this work are in course of publication; the paper is attached as
Appendix ii and the abstract is reproduced below.

"The Influence of Oxygen on Backscattering of Excited H when $H_1^+$, $H_2^+$ and $H_3^+$ Ions at 4 to 30 keV Energy are Incident on Molybdenum," by E. O. Rausch, M. W. Murray, H. Inouye and E. W. Thomas. J. Applied Physics (To be published).

A study has been made of the $H_8^-$ line intensity from excited H atoms scattered off Mo or Cu surfaces. Incident projectiles were 4 to 30 keV $H_1^+$, $H_2^+$, and $H_3^+$ ions. The integrated intensity of the atomic line emission increased as the ambient pressure of oxygen in the target chamber was raised from 10^-11 to 10^-6 torr. The experimental data suggested that this change of intensity is caused by a reduction in the radiation-less de-excitation of atoms by the Auger mechanism. A simplistic model was formulated which predicts the intensity as a function of ambient $O_2$ pressure, projectile beam current density and energy, but only in the case of $H^+$ ions on Mo. An attempt to extend the model to $H_3^+$ ions was not successful. For Cu targets an increase in the intensity as a function of oxygen pressure was also observed. However, no attempt was made to test the model in this case. At an incident angle of $60^\circ$ coefficients for sputtering of oxygen, $S_{Ox}$, adsorbed on Mo were determined to be $0.0025 \pm 0.0004$, $0.011$ and $0.017$ respectively, for 20 keV $H_1^+$, $H_2^+$, and $H_3^+$ projectiles.

The general implications for the MFE program is that the quantum state of low energy recoils, from a wall, will be a sensitive function of the wall condition, principally through changes to the Auger mechanisms that alter quantum state as the recoil emerges from the surface. Also we have measured a very high yield of sputtering of adsorbed gases by protons; which is of the order $10^{-3}$ atoms per ion for $60^\circ$ angle of incidence. For normal incidence (where yields are expected to be lower than at $60^\circ$) yields by Finfgeld for sputtering of clean bulk materials are of the same order as our measured yields for removal of adsorbed materials of monolayer, or less, concentrations.

In a subsidiary experiment we studied how excited state backscattering might be influenced by target temperature. Our previously published work

was all at room temperature while any practical MFE device will be at a wall temperature of 800°C or more. Moreover there is unpublished data from elsewhere\textsuperscript{6} indicating a temperature dependence; such a temperature dependence is at complete variance with the model we have been using to represent excited state formation. We have repeated many of our previous measurements of excited state formation with the use of elevated target temperatures to 400°C; no temperature dependence was observed. We do plan equipment modifications to permit raising temperature to 800°C for further tests but the indication at present is that the backscattering and neutralization of ions at metal surfaces is not temperature dependent.

We have been concerned as to whether our successful modelling of backscattering was in some way peculiar to the case of simple neutral recoils such as H and He. To provide a test of more general applicability we studied excited state formation in the case of B\textsuperscript{+} ions incident on metals leading to formation of excited B and B\textsuperscript{+}. The general conclusion is that the model works equally well for these recoil species. The data are in course of publication, the paper is reproduced as Appendix iii and the abstract is given below.

"Excited State Formed by B\textsuperscript{+} Impact on Metallic Cu and Mo," by E. W. Thomas, H. Inouye, and E. O. Rausch.

Impact of 60 to 200 keV Be\textsuperscript{+} ions on polycrystalline Cu and Mo targets gives rise to emission from backscattered excited Be and Be\textsuperscript{+}. The emission coefficients for these lines are presented as a function of impact energy and analyzed to provide a measure of the survival parameter, A/a, for the atom or ion recoiling from the target. For B recoiling from both Cu and Mo this is found to be 1.3 x 10\textsuperscript{8} cm/sec while for recoiling B\textsuperscript{+} it is 2.4 x 10\textsuperscript{8}. In the case of recoiling B\textsuperscript{+} the radiationless de-excitation must be by the Auger mechanism. Emission coefficients for the 3247, 3274 Å CuI doublet are also presented and rise slowly throughout the energy range studied.

\textsuperscript{6} R. C. Amme, University of Denver (Private communication).
C. Evaluation of the SCANIIR Technique of Surface Analysis

In principle one may study the composition of a surface by analyzing the optical emission spectrum induced by impact of a heavy ion beam on a surface. Most of the emission is from sputtered surface atoms and the intensity of a spectral line is related to the concentration of the element involved. The technique is known as SCANIIR (Surface Composition Analysis by Neutral and Ion Induced Radiation). In many respects SCANIIR is similar to SIMS; in the latter case one records sputtered charged particles. SCANIIR has been considered as a possible technique for insitu analysis of MFE device walls. Most conventional surface analysis techniques (Auger, SIMS, LEED etc) cannot be operated within the environment of the MFE device. By contrast SCANIIR may use a neutral probing beam, relies on photons as an output signal, and the sensitive detection equipment may be placed at a considerable distance from the wall under study; thus SCANIIR can operate directly on the wall of the MFE device while other techniques must have a segment of wall removed for analysis elsewhere.

We have evaluated the potential usefulness of SCANIIR by analyzing the visible and near u-v emission from metal samples onto which H, C, or O have been absorbed, or into which B, C, N, Cu, or Fe have been implanted at low energies. The general conclusion is that H and O cannot be detected at visible wavelengths, C and N can be detected with difficulty and only metals on the surface can be reliably and readily detected. These conclusions are generally in accord with those of other scientists. It is sometimes suggested \(^7\) that available signals can be enhanced by exposing the surface to oxygen permitting even H to be detected. We consider this not to be useful for MFE since it is obviously impractical to flood the device with \(O_2\) for the purpose of analyzing the wall situation.

We believe that SCANIIR is viable only if the optical emissions utilized

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7. I. Tsong, Pennsylvania State University (Private Conversation).
are transitions to the ground state; these are the most intense. For H, C, and O these lie in the vacuum uv. Private communications from Schartner (University of Giessen) show that some success has been achieved and we shall be testing this within one month.

Full details of our evaluation to date are given in Appendix iv. They include a demonstration that the density of chromium at steel surface exceeds that of the bulk; also an attempt to depth profile Cu implanted in C which indicates how Cu, being insoluble, migrates into the carbon, probably by radiation enhanced diffusion. The SCANIIR technique is useful for detection of metals and we anticipate shortly demonstrating utility for H, C and O when detection at 1000-2000 Å wavelengths is utilized.

D. Backscattering from Surfaces - Total Particle Fluxes, Charge States and Energy Distributions

This experiment seeks to measure directly the flux of recoiling hydrogen atoms and the distribution of this flux with velocity, charge state and recoil energy. The ultimate objective is to bombard with H⁺ and to measure directly the angular and energy distributions of the scattered H⁺, H⁰ and H⁻. This is a very difficult experiment to perform, particularly in the monitoring of the low energy recoils. A german group has produced most of the available data on this phenomena; the fact that their data for low energy recoils has changed² by an order of magnitude from their first publication³ is testimony to the difficulty of the problem. Our work on this subject has proceeded more slowly than expected; an interim report on certain measurements is included here as Appendix v.

Protons of energy 1 to 30 keV are directed onto a metal target. Particles scattered into a limited range of angles are selected by a pair of slits; the transmitted flux is electrostatically analyzed and the different charged components monitored either as currents or by single particle detectors. The electrostatic analyzer may be used to analyze the energy distribution of the charged particles; the neutrals can be stripped in a gas cell and then energy analyzed as charged particles.

In Appendix v we present a sample energy spectrum of $H^+$ recoiling from Cu. The peak intensity (for 15 keV incident ions) is at a recoil energy of about 8 keV for the case of $H^+$ ions incident at $20^\circ$ to the surface and recoiling at $10^\circ$ to the surface. By contrast, work of the German group for much larger angles of incidence and emergence, shows the recoil flux to peak at 2 keV. There are clearly substantial differences between normal and grazing incidence.

We also present in Appendix v for the first time a direct absolute measure of the backscattered ion flux; previous measurements by others involve relative measurements with absolute magnitude assigned by normalization procedures. We provide also estimates of the total backscattered flux which lie a factor of three below theoretical predictions.

E. Related Work in this Laboratory

Under sponsorship of NSF we are studying chemical reactions induced by ion impact on surfaces. We utilize the optical emission spectrum to indicate the presence of a particular radicle, and, in combination with Auger monitoring of atomic concentrations, we are to deduce reaction rates for formation of the radicle. In brief the problem being considered are as follows:
(1) The formation of CN (detected by the CN vibrational spectrum around 3000 Å) induced by heavy ion impact onto an alkalie halide surface carrying an adsorbed layer of CO, H₂O and N₂. In this case the surface appears to catalyze the reaction.

(2) The formation of CH (detected by the CH emission bands) due to sequential implantation of C and H into silicon or implantation of H into C. This represents the reaction of C and H to form hydrocarbons and is obviously related to the chemical sputtering of carbon which seems to preclude use of carbon liners in MFE machines.

(3) The formation of N₂ (detected by N₂ emission bands) due to high dose N⁺ implantation into Si. Presumably the implanted N is exposed by sputtering of the substrate and recombines to form N₂ before being removed by collision cascades.

(4) The formation of silicon nitride due to implantation of N⁺ into Si. This again represents a case where the implanted species combines chemically with the substrate.

These various projects are of peripheral interest to MFE in that they represent changes to surface chemistry due to ion impact. This is a subject of major concern to the long term development of MFE since there is little known on the chemical behavior of materials in the extreme environment of the plasma device. No details of these projects are presented here since they have been pursued only for a couple of months.

V. Participation on ERDA Task Group

During the course of the contract year the project Director, E. W. Thomas, participated in the work of the "Task Group on Plasma Material Interaction." His function was a member of the sub-task group considering "Desorption, Chemical Effects and H. Interactions." Considerable time was devoted to this work,
including meetings at Argonne and Sandia. The results of this work are now being coalesced into an ERDA report.

VI. Conferences and Other Travel

During the period covered by this report, Dr. Thomas attended the following conferences and gave presentations based in part on the work performed under this contract.


(b) APS Meeting in San Diego, March 21, 1977. An invited paper was presented entitled "Formation of Excited States by Ion Impact on Surfaces."

(c) "Third International Conference on the Application of Ion Beams to Surface Analysis." Washington, D.C., June 27, 1977.

(d) "First International Conference on Low Energy Ion Beams," University of Salford (U.K.) 5-8 September 1977.


The project director also made visits to the following research establishments:

(i) Argonne National Lab.

(ii) Oak Ridge National Lab.

(iii) Sandia Corp.

(iv) FOM Institute (Amsterdam, The Netherlands)

(v) Culham Laboratory (England)

(vi) Harwell Laboratory (England)

(vii) Kutchartov Institute (Moscow, USSR)

(viii) McMaster University (Canada)
In all cases the visits were for the purpose of conferring with scientists active in the fusion program (or related areas) in the USA and abroad.

VII. Personnel

Professor E. W. Thomas has been Project Director and Principal Investigator in this work. He has devoted to it 15% of time in the academic year and 50% of full time during the summer.

Dr. E. O. Rausch worked on this project as a postdoctoral Research Assistant for half full time until July 1977. At that time he was replaced by K. O. Legg who continues to occupy that position, devoting one half time to the project.

Physics graduate students who have worked on this project include J. E. Harriss, H. Inouye, W. A. Metz, and A. J. Senol.

VIII. Publications

Four articles have been published and six have been accepted for publication. These are fully listed as Appendix (vi), in the format requested by ERDA.

XI. Appendixes

(i) "Excited H Formation by 2-200 keV H_1^+, H_2^+ and H_3^+ Ion Impact on Metal Surfaces. E. O. Rausch, H. Inouye, A. J. Senol and E. W. Thomas, Phys. Rev. A. (Accepted for publication).

(ii) "The Influence of Oxygen on Backscattering of Excited H when H_1^+, H_2^+, and H_3^+ Ions at 4 to 30 keV Energy are Incident on Molybdenum. E. O. Rausch, M. W. Murray, H. Inouye, and E. W. Thomas. J. Appl. Phys. (Accepted for Publication).

(iv) "Analysis of Surfaces by the SCANIIR Technique (a preliminary report).

(v) "Backscattering of 1-30 keV Hydrogen from Metal Surfaces" by J. E. Harriss (a preliminary report).

(vi) Listing of Publications (ERDA Form).
Excited H Formation by 2-200 keV \( \text{H}^+, \text{H}_2^+, \text{and H}_3^+ \) Ion Impact on Metal Surfaces

E.O. Rausch, H. Inouye, A.J. Senol, and E.W. Thomas
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332

Abstract

The coefficient for emission of the Balmer \( \beta \) line from backscattered H atoms has been measured for 2-200 keV hydrogen ion impact on polycrystalline targets of Mo, Cu and stainless steel. The dependence on primary energy is successfully modelled using a modification of a formulation used previously at low (2 to 30 keV) impact energies.
When an energetic hydrogen ion beam is incident on a metal surface a small fraction of the projectiles will backscatter as excited hydrogen atoms. These atoms may lose their excited electron by radiationless mechanisms during interaction with the surface or they may recede beyond the interaction region and subsequently emit photons by normal radiative decay. The integrated photon intensity in a spectral line is related to the backscattered flux of excited atoms whereas the line profile yields information on the distribution on speed and angle of the particles. The observed spectral lines are Doppler-broadened, because most particles escape from the surface with an energy greater than 1 keV. A model which can predict the backscattered flux of all particles (independent of charge or excited state) was developed by McCracken and Freeman and utilized by Baird et al. to predict backscattering of excited particles for impact energies to 30 keV. It was assumed that the projectile penetrating a target undergoes a single large angle scattering event and returns to the surface, while in the target the particle loses its energy by electronic stopping. Because of variations in energy loss, penetration depth and scattering angle the backscattered flux will exhibit a velocity distribution. By summing the contribution to the backscattered flux over all possible velocities of the emergent particle it is possible to predict the integrated intensity of emission in a particular transition. Equation 1 has been used to predict, $\gamma_j$, the number of backscattered atoms in the excited state $j$ per incident ion.

$$\gamma_j(E_o, \phi) = \int_{v_f}^{v_u} f(v, E_o, \phi) F_j(v) \exp\left(-\frac{\Lambda}{a v_f}\right) dv$$

Here $E_o$ is the incident particle energy and $\phi$ the angle of incidence. The factor $f(v, E_o, \phi) \cdot dv$ represents the flux of backscattered particles emerging.
with a velocity component perpendicular to the surface between $v_x$ and $v_x + dv_x$; this can be obtained from the predictions of McCracken and Freeman. The factor $P_j$ is the probability that the backscattered particle is in an excited state $j$; this may be written as a product between, $P_o$, the fraction of recoils which are neutral and, $P_j$, the fraction of neutrals which are excited (i.e. $P_j = P_o P_j$). The exponential factor in Eq. 1 is explained in our earlier work and accounts for the loss of excited atoms by radiationless decay mechanisms; it has been suggested that the decay mechanism is by an Auger transition and the constants $A$ and $a$ are related to the height and width of the potential barrier between the recoiling atom and the solid. In performing the integration of Eq. 1 we use an upper limit of velocity ($v_u$) that corresponds to projectile scattering from a surface atom without penetration and we adopt a lower velocity limit corresponding to an emergent energy of 20 eV; below this lower limit we presume that projectiles are trapped in the lattice and do not recoil.

Strictly speaking Eq. 1 predicts the formation of an excited state $j$ while the observations are of emission in the transition $j \rightarrow k$. We have however shown that cascade population of $n=4$ states of hydrogen is negligible so that the measured emission coefficient $\gamma_{jk}$ (Balmer $\beta$ photons per ion incident) is proportional to the predicted excitation coefficient $\gamma_j$ (reflected atoms in the $n=4$ state per ion incident). In our previous work we have made relative measurements of $\gamma_{jk}$, as a function of impact energy $E_o$ in the range 5 to 30 keV and fitted $\gamma_j$ from Eq. 1 to the data, permitting us to derive a value for the ratio $A/a$; the assumption was made that $P_j$ was a constant.

For targets of Cu, Mo and type 304 stainless steel the value of $A/a$ was determined to be $1.7 \times 10^8$ cm/sec with an accuracy of $\pm 25\%$.

The objective of the present work was to extend the previous studies (limited to a maximum impact energy of 30 keV) up to impact energies of 200 keV. It was to be anticipated that at high energies the factor $P_j$ will be
energy dependent and the assumption of a constant $F_j$ made in earlier work will be invalid. Thus, this present extended study will permit a determination of how the formation of backscattered excited atoms varies with impact energy, and might provide a test of recent theoretical predictions.

The only substantive change in experimental details from our previous work\(^3\) has been the use of a different ion accelerator to increase the projectile energy from the previous upper limit of 30 keV to the present limit of 200 keV. Briefly, the accelerated ions strike the target at an incident angle of 60° (angle between the ion beam and target normal). An optical monochromator views the target at 90° with respect to the ion beam. Wide entrance and exit slits were employed on the monochromator to maximize signal to noise ratio; this resulted in a rather poor resolution (48°). We scanned through the Doppler-broadened Balmer $\beta$ line and integrated the area under the peak. All values of $\gamma_{jk}$ reproduced here are relative values; absolute measurements of $\gamma_{jk}$ have been presented in earlier work.\(^4,5\) The integrated line intensity as a function of incident projectile energy is shown in Figs. 1, 2 and 3 for targets of Mo, Cu and type 304 stainless steel respectively. Data at energies less than 30 keV were reported earlier, but are also reproduced here. We used not only $H_1^+$ but also $H_2^+$ and $H_3^+$ projectiles, because it was previously shown that at low ambient oxygen pressures ($\nu10^{-11}$ torr) in the target all $\gamma_{jk}$ for $H_2^+$ and $H_3^+$ with energy $E$ is the same as to $\gamma_{jk}$ for $H_1^+$ with energy $E/2$ and $E/3$, respectively.

The intention was to model the data using Eq. 1 to predict $\gamma_j$ and to compare it with experiment assuming that this also represented the relative behavior of the measured emission coefficient $\gamma_{jk}$. The velocity distribution of backscattered particles, $f(v, E, \phi) dv$ was to be obtained from the formulation of McCracken and Freeman.\(^2\) This formulation is strictly valid only for those incident velocities less than the velocity of an electron.
in the first Bohr orbit where the LSS theory of electronic stopping is valid; this limits applicability to protons of less than 25 keV incident energy. However McCracken and Freeman showed that the formulation agreed very well with direct experimental measurements of backscattering with 120 keV incident protons. Therefore we have continued to use that prediction here.

In the analysis of earlier experiments at lower impact energies (<30 keV) we have assumed that the probability of a recoil being an excited neutral, $F_e$, was independent of recoil velocity. This is not likely to be correct at the higher energies of the present experiment and we have examined various energy dependent forms. The factor $F_e$ can be written as the product $P_o P_j$, where $P_o$ is the probability of the recoil being a neutral and $P_j$ is the probability of the neutral recoil being excited. There are two recent theoretical predictions of $P_o$. One is by Cross and considers the projectile to be undergoing successive processes of charge transfer neutralisation and stripping in the solid. The other is by Kitagawa and Ohtsuki; it considers the projectile to remain stripped in the solid and allows neutral formation by an electron recombination mechanism as the projectile escapes from the surface. Unfortunately, the success of Cross's theory is confined to carbon targets and to particles with exit energies $\geq 100$ keV. The predictions of Kitagawa et al. are for Au and differ from the experimental data by up to 70% within the energy range 30 keV to 200 keV, although at energies below 30 keV the predictions agree with experiment. Since neither prediction adequately covers the range of our experiment we decided instead to use the experimental measurements of $P_o$ for $H^+$ on Mo and Cu by Behrisch et al. and to extend their range by using the low energy approximation of Kitagawa et al. at energies $\leq 20$ keV and the high energy approximation of Cross at energies $\geq 150$ keV. It would appear that the value of $P_o$ is not a sensitive function of material. The experimental data of Behrisch et al. for Mo and Cu are within 20% of experimental data for C, Au, Cr, Fe and Al by Chateau-Thierry et al.; the theoretical
values by Cross \(^8\) for C are similar to the theoretical values of Kitagawa and Ohtsuki \(^9\) for Au. Consequently we feel that our use of experimental data with extrapolations from theory is quite reliable. For completeness we reproduce in Fig. 4 the actual values of \(P_o\) that we employed. Both the theory of Cross \(^8\) and that of Kitagawa and Ohtsuki \(^9\) suggest that the probability of a neutral being in an excited state, \(P_j\), is inversely proportioned to the cube of the principal quantum number, we assume that relationship here. Thus the energy dependence of \(F_j\) \(\left(=P_o P_j\right)\) is given in relative terms simply by the variation of \(P_o\).

In Fig. 1 we show a variety of computed excitation coefficients, \(\gamma_j\) (from Eq. 1) normalized to the experimental emission coefficients \(\gamma_{jk}\); both computed and experimental data are relative values. One computed curve employs a constant \(F_j\) and a value for \(A/a\) of \(1.7 \times 10^8\) cm/sec; these are the conditions used in our earlier analyses \(^1\) for data at energies below 30 keV. As expected there is a discrepancy between computation and experiment at high energies. A second computation employs the same survival parameter \(A/a\), but an energy dependent \(F_j\) obtained by the manner described above. In this case the predictions of the model agree well with experiment. For completeness a computation is also shown with the \(F_j\) derived as described above and the survival parameter \(A/a\) adjusted to give the best fit to experiment. In this case the value of \(A/a\) derived by fitting to the data comes out as \(1.9 \times 10^8\) cm/sec; within the accuracy we assign \(^1\) to \(A/a\) \((\pm 25\%)\) this is not significantly different from the previously derived \(^1\) value \((1.7 \times 10^8\) cm/sec). We have also utilized Kitagawa and Ohtsuki's prediction of \(P_o\) and find the predicted \(\gamma_j\) to be less than 10% different from the predictions using \(P_o\) from Fig. 4; this prediction is not shown to avoid confusion with the other lines on Fig. 1.

In Fig. 2 we show a single prediction for the Cu target utilizing the neutral
formation probability $P_o$ from Fig. 4 and a survival coefficient $A/a$ of $1.9 \times 10^8$ cm/sec; again the prediction agrees with measurement within experimental accuracy. We show no predicted $\gamma_j$ for the case of stainless steel in Fig. 3 since there is no experimental data for $P_o$ on which to base the model; the line on that figure is drawn freely to indicate only the general trend of the experimental data.

Some measurements of Doppler-broadened line shape were performed and compared with the predicted shape from the model in the manner of our earlier work. These results are not presented in detail since they were taken with a poor spectral resolution (to enhance signal strength) and therefore are dominated by the instrumental line width. As an example the width of the line at half maximum intensity for 180 keV H$^+$ on Mo was measured to be $75^\circ$ using the instrumental resolution of $48^\circ$. The predicted width using the model with $P_o$ from Fig. 4 and $A/a=1.9 \times 10^8$ cm/sec with allowance for instrumental line width is $77.1^\circ$; this is in satisfactory agreement within the experimental accuracy.

By contrast the predicted width assuming probability of excited state formation is not dependent on energy (i.e. $F_j=constant$) is $87.7^\circ$. Similar figures are found for the Cu target. Thus the line shape is consistent with the adopted model.

The present work has extended the previous measurements to higher impact energies and demonstrated that the model adopted for predicting emission coefficient remains valid provided account is taken of the energy dependence of the probability of recoils being excited, $F_j$. It is clear from Fig. 1 that the emission coefficient is not a sensitive function of $F_j$; assumption of a constant $F_j$ at all energies overestimates the emission coefficient by only a factor of two at 200 keV. The emission coefficient for a high impact energy proton is dominated by the low energy recoils which constitutes the greatest part of the recoil energy spectrum.

The predictions of the model are not significantly altered as one changes
the adopted value of $P_0$ from the experimental data of Fig. 4 to the predictions of Kitagawa and Ohtsuki and finally to those of Cross. Although the two theoretical predictions are based on entirely different concepts, their predictions are sufficiently close that they agree equally well with our experiment. Thus the present experiment does not provide a test as to which prediction is most accurate.

Our earlier success in modelling emission coefficients at low energies with a constant $F_j$ is explained by the insensitivity of the model results to the form of $F_j$ adopted.
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References

Figure Captions

1. Projectile energy dependence of Balmer $\beta$ line emission coefficient $\gamma_{jk}$ for hydrogen backscattered from an Mo target. The data points are experimental measurements; the lines are predicted values from the model normalized to experiment at 30 keV. The dashed line is for $F_j$ constant and $\Lambda/a = 1.7 \times 10^8$ cm/sec; the dotted line is for an energy dependent $F_j$ and $\Lambda/a = 1.7 \times 10^8$ cm/sec; the solid line is for an energy dependent $F_j$ and $\Lambda/a = 1.9 \times 10^8$ cm/sec.

2. Projectile energy dependence of Balmer $\beta$ line emission from backscattered $H^*$ atoms for a Cu target. The solid line indicates theoretical values with $\Lambda/a = 1.9 \times 10^8$ cm/sec and $F_j$ is dependent on emergent particle energy.

3. Projectile energy dependence of Balmer $\beta$ line emission from backscattered $H^*$ atoms for a stainless steel target (SS304). The dashed line indicates the trend of the data.

4. The values of neutralization probability $P_0$ assumed in the model. These are determined from the experimental data of Behrisch et al.\textsuperscript{10} extrapolated to low energies by the theory of Kitagawa and Ohtsuki\textsuperscript{9} and to high energies by the theory of Cross.\textsuperscript{8}
INTENSITY PER NUCLEON
(ARBITRARY UNITS)

PROJECTILE ENERGY (keV PER NUCLEON)
FRACTION OF NEUTRAL PARTICLES

ENERGY OF EMERGING PARTICLES (keV)

- $H_1^+$ ON Mo
- $H_1^+$ ON Cu
- KITAGAWA ET AL.
The influence of oxygen on backscattering of excited H when H\textsuperscript{+}, H\textsubscript{2}, and H\textsubscript{3} ions at 4–30 keV energy are incident on molybdenum\textsuperscript{a)}

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A study has been made of the H\textsubscript{\beta} line intensity from excited H atoms scattered off Mo or Cu surfaces. Incident projectiles were 4–30-keV H\textsuperscript{+}, H\textsubscript{2}, and H\textsubscript{3} ions. The integrated intensity of the atomic line emission increased as the ambient pressure of oxygen in the target chamber was raised from 10\textsuperscript{-11} to 10\textsuperscript{-6} Torr. The experimental data suggested that this change of intensity is caused by a reduction in the radiationless deexcitation of atoms by the Auger mechanism. A simplistic model was formulated which predicts the intensity as a function of ambient O\textsubscript{2} pressure, projectile beam current density, and energy, but only in the case of H\textsuperscript{+} ions on Mo. An attempt to extend the model to H\textsubscript{2} ions was not successful. For Cu targets an increase in the intensity as a function of oxygen pressure was also observed. However, no attempt was made to test the model in this case. At an incident angle of 60° coefficients for sputtering of oxygen, S\textsubscript{ox}, adsorbed on Mo were determined to be 0.0025±0.0004, 0.011, and 0.017, respectively, for 20-keV H\textsuperscript{+}, H\textsubscript{2}, and H\textsubscript{3} projectiles.

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I. INTRODUCTION

The impact of 4–30-keV H\textsuperscript{+}, H\textsubscript{2} ions on metal targets results in the formation of backscattered excited H atoms. These subsequently decay by photon emission which can be observed as Doppler-broadened spectral lines. Information on the distribution in speed and angle of the scattered particles can be extracted from the line profile.\textsuperscript{1–3} The integrated line intensity represents the flux of backscattered atoms in the relevant excited state.

In previous work\textsuperscript{3} it was shown that the flux of backscattered atoms may be given by the following:

\[
\gamma_j(E_0, \phi) = \int_{V_1}^{V_2} f(V_i, E_0, \phi) F_j \exp(-A/a V_i) dV_i. \tag{1}
\]

Here \(\gamma_j\) is the backscattered flux in excited state \(j\) per incident nucleon, for an incident energy of \(E_0\) and angle of \(\phi\). \(f(V_i, E_0, \phi) dV_i\) is the flux of backscattered particles emerging with velocity components perpendicular to the surface between \(V_1\) and \(V_2 + dV_i\). \(F_j\) is the fraction of backscattered atoms formed in the state \(j\) and the exponential factor accounts for the loss of excited atoms by radiationless decay mechanisms. It was suggested that the Auger effect\textsuperscript{1,2} is the relevant nonradiative decay mechanism; in this case the constants \(A\) and \(a\) are related to the height and width of the potential barrier between the recoiling atom and the solid. The effect of a large \(A/a\) (\(\sim 10^7\) cm/sec for Mo) is to allow only fast particles to escape from the target in an excited state. The resultant line shape is broadened and is made up of two components, a sharp blue-shifted peak and a small red-shifted contribution (see Fig. 1). Both the blue- and red-shifted photons originate from H atoms having a velocity component that is directed either toward or away from the observer, respectively.

In the present work we investigated how the backscattered excited flux from a Cu or Mo target is influenced by the presence of oxygen. First, we will show that the \(A/a\) coefficient in Eq. (1) differs depending on whether the target is clean or covered with oxygen. Based on this change in \(A/a\) we will then formulate a model which will relate the photon emission from the backscattered flux of excited atoms to the oxygen surface coverage.
II. EXPERIMENTAL

The experimental arrangements remain unchanged from our previous work\textsuperscript{1-3} and will not be described in detail. An ion beam (energy 4–30 keV) of \( \text{H}_2^+ \), \( \text{H}_2 \) and \( \text{H}_3^+ \) projectiles is incident on a metal target and the induced light emission is observed by an optical monochromator viewing through a spahire window. The optical axis is perpendicular to the ion beam and the angle of incidence (measured between the incident beam and the surface normal) can be varied from 0° to 90°. Most measurements presented here are of the integrated line intensity of the \( n = 4 \rightarrow n = 2 \) transition in \( \text{H}_2 \); this was performed directly by setting the spectrometer resolution to receive the whole Doppler-broadened line. We note that all values of \( \gamma \) presented in this work are relative values.

The composition of gas in the target region could be analyzed and measured with a partial pressure analyzer (Ultek Model 607). Typical base pressures were \( 1 \times 10^{-6} \) Torr. A leak valve was provided so that oxygen gas could be deliberately introduced into the target chamber to pressures up to \( 10^{-6} \) Torr. In this way we were able to form adsorbed gas layers on the target surface. At a typical base pressure of \( 10^{-6} \) Torr the oxygen partial pressure was only \( 5 \times 10^{-11} \) Torr. The area of the target irradiated by the ion beam was approximately 5 mm\(^2\) and the beam current density varied from 0 to 140 \( \mu \text{A/cm}^2 \). The target materials were polycrystalline metal foils of Mo and Cu obtained from the Materials Research Corporation. The purity of the metals was specified to be better than 99.9%.

III. RESULTS AND DISCUSSION

A. Line-shape analysis and energy dependence

In previous work\textsuperscript{1-3} the \( A/\alpha \) coefficient was extracted from the data either by analysis of the line shape or by analysis of the energy dependence of the total line intensity. In either case a suitable \( A/\alpha \) will permit good agreement between predictions and data. For this work we used both methods to determine the change in \( A/\alpha \) associated with adsorption of oxygen on the target surface. It is suggested that the adsorbed oxygen would alter the potential barrier between the backscattered atom and the target surface; this should cause a change in the \( A/\alpha \) value. A decrease in \( A/\alpha \) would allow the slower hydrogenic particles to escape from the surface without losing their excited electrons by means of the Auger mechanism. This increases the probability that the particles will escape in an excited state. Hence, an increase in the integrated intensity of the line shape and a narrowing of the line profile is expected with the peak intensity occurring closer to the unshifted wavelength. On the other hand, an increase in \( A/\alpha \) would have the opposite effect.

We shall first investigate the energy dependence of the integrated \( \text{H}_2^+ \) line intensity for \( \text{H}_2^+ \) on Mo at an incidence angle of 60°. Figure 2 shows the experimental data (circles) and the predicted intensities (solid lines) at \( 10^{-11} \) and \( 10^{-6} \) Torr oxygen pressure. The predictions are obtained from Eq. (1). Since \( F \) is unknown, we have simply normalized the predicted value to experiment at 22 keV on line B; the factor \( A/\alpha \) in Eq. (1) was then adjusted to provide a good fit of the predicted \( \gamma \) to experiment. It was shown earlier\textsuperscript{4} that the predicted \( \gamma \) is very sensitive to the choice of \( A/\alpha \). By this procedure we deduced that \( A/\alpha = 1.60 \times 10^6 \text{ cm/sec} \) is appropriate for fitting the data at \( 10^{-11} \) Torr and \( A/\alpha = 1.22 \times 10^6 \text{ cm/sec} \) is appropriate at \( 10^{-6} \) Torr oxygen pressure. The precision with which the \( A/\alpha \) values could be determined in this manner was \( \pm 0.05 \times 10^6 \text{ cm/sec} \). Hence, the difference in the \( A/\alpha \) values is given by \( (0.38 \pm 0.1) \times 10^3 \text{ cm/sec} \).

Next we calculated the amount by which the \( A/\alpha \) coefficient must change from its value at \( 10^{-11} \) Torr oxygen pressure to account for the increase in integrated photon intensity at 20 keV when the oxygen pressure was changed to \( 10^{-6} \) Torr. Figure 3 shows the integrated intensity of the \( \text{H}_2^+ \) transition (measured as signal per
unit beam current) as a function of ambient O_2 pressure in the target cell for two different beam currents. The observed difference in intensity between 10^{-11} and 10^{-6} Torr oxygen pressure is consistent with a change in A/a from the clean surface value of 1.6 x 10^{16} cm^2/sec (at 10^{-11} Torr) to a value of 1.29 x 10^{16} cm^2/sec for the conditions at 10^{-6} Torr pressure of O_2. This in turn should alter the atomic line by shifting the peak intensity by 0.97 Å to lower wavelengths. The line shape was studied with a resolution of 8 Å for the condition of 10^{-6} Torr O_2 as shown in Fig. 1. Its peak position was shifted by 2 ± 1 Å to lower wavelengths, but the profile itself remained unchanged. Thus, the difference in A/a values derived from the energy dependence is in good agreement with that derived from the changes in integrated atomic line intensity at a single energy within the precision stated earlier. The apparent difference between the A/a value determined for this work (1.6 x 10^{16} cm^2/sec at 10^{-11} Torr) and that determined previously^1 (1.8 x 10^{16} cm^2/sec at 10^{-6} Torr) can be explained on the basis of the limited accuracy of these earlier values which were reported^1 to be reliable only to within ±25%. The agreement between predicted and observed energy dependence in Fig. 2 is surprisingly good in view of the fact that only A/a was varied. This comparison suggests that the factor F_i in Eq. (1), the probability of a scattered particle being formed in a specific excited state, is not significantly affected by the presence of oxygen on the surface of the target. Therefore, we base the subsequent formulation of our model on the assumption that a change in A/a is the mechanism responsible for the oxygen effect.

B. Pressure dependence of intensity

We propose the following qualitative model to explain the results in Figs. 1–3. A clean target surface produces a certain photon emission coefficient γ(A/a) governed by the constants A and a appropriate to the Auger effect at a clean surface. At high oxygen pressures, the surface is completely covered with adsorbed oxygen causing a change of the constants to A' and a' with a consequential new emission coefficient γ(A'/a'). At intermediate conditions a fraction f_{ox} of the surface is covered with oxygen and the resultant photon emission per incident nucleon, Γ, is given by the following weighted sum:

\[ Γ = \frac{I}{n I_p} = γ \frac{A}{a} (1 - f_{ox}) + γ (A'/a') f_{ox}, \]  

(2)

where I is the integrated atomic line intensity, I_p is the beam current density, and n is the number of nucleons per incident ion (n is unity for H and 2 for H^2). This involves the assumption that the resultant intensity consists of recoils which interact with a fraction f_{ox} of oxygen-covered surface and a fraction (1 - f_{ox}) of metal surface. At sufficiently low pressures the target is presumed to be atomically clean and f_{ox} = 0; at high pressures the target is covered with oxygen and f_{ox} is presumed to be equal to unity.

In order to express f_{ox} in variables which can be determined experimentally we will first consider the time dependence of the density of oxygen on the target surface. The oxygen surface coverage is related to the rate of arrival of atoms from the surrounding gas (P_{ox} atoms/cm^2/sec) and the sticking probability in relation to the rate of oxygen removal by the ion incident beam of flux I_p (ions, cm^2/sec) and oxygen sputtering coefficient S_{ox} (atoms removed ion incident for f_{ox} = 1). Van der Weg et al. have shown that the concentration of oxygen is given by

\[ \frac{dN_{ox}}{dt} = -\left( \frac{I_p S_{ox} N_{ox}}{N_{Mo}} + P_{ox} \right) \alpha, \]  

(3)

where N_{ox} is the atomic surface density of the metal. The sticking coefficient can be taken as unity for f_{ox} = 0; however, for high values of f_{ox} the coefficient α must obviously become small. A functional form of α which allowed us to fit the measured data is given by

\[ α = 1 - f_{ox}, \]  

(4)

where f_{ox} = N_{ox}/N_{Mo}. The physical interpretation of Eq. (4) is as follows. We assumed that under ion bombardment conditions an oxygen molecule will stick to a Mo atom, but not to an oxygen atom on the surface of the target. As a result, the sticking probability α is proportional to the surface area of the target which is not covered by oxygen (i.e., 1 - f_{ox}).

We are now able to obtain a solution to Eq. (3) which is given by

\[ f_{ox} = \frac{N_{ox}}{N_{Mo}} = \left( \frac{\ln S_{ox}}{P_{ox} + 1} \right) - \left( \frac{C}{I_p S_{ox} + P_{ox}} \right) \exp \left[ -\left( \ln S_{ox} + P_{ox} \right) / N_{Mo} \right], \]  

(5)

Here, C is a constant of integration. After a sufficiently long time has elapsed an equilibrium condition between removal and adsorption of oxygen will have been established. The exponential term in Eq. (5) will become negligibly small and the fractional surface coverage f_{ox} is then given by Eq. (6).

\[ f_{ox} = \left( \frac{2 \ln S_{ox} k T}{P V} + 1 \right)^{-1}. \]  

(6)

Here, P_{ox} has been related to the partial pressure of oxygen P by

\[ P_{ox} = 2 (P V / 4 k T) = P V / 2 k T, \]  

(7)

where V is the mean thermal velocity of the oxygen molecules, k is Boltzmann's constant, and T is the temperature in degrees Kelvin. The factor 2 outside the parenthesis in Eq. (7) was included to allow for the fact that oxygen is diatomic. The complete formula which predicts the intensity as a function of oxygen pressure, beam current density, and energy can now be derived by substituting Eq. (6) into Eq. (2), which yields

\[ Γ = \frac{I}{n I_p} = γ \left( \frac{2 \ln S_{ox} k T}{P V} + 1 \right)^{-1} \left( γ' - γ \right), \]  

(8)

where γ = γ(A/a) and γ' = γ(A'/a').
With this model it follows that the measured values of \( \Gamma \) at different beam currents should be a family of curves tending asymptotically to the same values at high \( f_{\text{ox}} \) (i.e., high pressure where \( f_{\text{ox}} = 1 \)) and low \( f_{\text{ox}} \) (i.e., low pressure where \( f_{\text{ox}} = 0 \)). This is in fact consistent with Fig. 2. Data for curves A and B were taken with beam currents of 30 \( \mu \)A/cm\(^2\) (curve A) and 140 \( \mu \)A/cm\(^2\) (curve B). The data fitting procedure was carried out as follows. The constants \( \gamma \) and \( \gamma' \) were extracted from the data at 10\(^{-11}\) and 10\(^{-6}\) Torr oxygen pressure, respectively. (Here we would point out that \( \gamma' = 1.52 \) for 20 keV H\(^+\) impact at 60° incidence angle.) \( \Gamma \) was set to the room-temperature value of 300 K and \( f_B \) was set to the higher beam current density (140 \( \mu \)A/cm\(^2\)). The predictions were then fitted to curve B by adjusting the sputtering coefficient \( S_{\text{ox}} \) to 0.0029. Thereafter, \( S_{\text{ox}} \) remained constant and \( f_B \) was changed to the lower beam current density (30 \( \mu \)A/cm\(^2\)) to predict curve A. We note that we tried to use other functional forms for \( \sigma \) as well. For example, we approximated, up to one monolayer (2 \( \times \) 10\(^{15}\) atoms), the sticking probability profile for oxygen on Mo reported by Horgan and King\(^\text{a}\) (their Fig. 9). However, this form was not successful in modeling our data.

The pressure dependence of \( \Gamma \) was also investigated at an incidence angle of 15° (see Fig. 4). In this case, the model predicted a ratio for \( \gamma' / \gamma \) equal to 1.46. By lowering the sputtering coefficient to a value of 0.0012 we found that the predictions were again consistent with the data within experimental precision. It should be noted that due to the low sputtering coefficients at near-normal incidence the value of \( \Gamma \) does not reach its asymptotic limit at the lowest oxygen partial pressures obtainable in our equipment.

We also observed an increase in the H\(_2\) line intensity with oxygen pressure when Cu was bombarded by H\(^+\) ions at an incidence angle of 60° (Fig. 5). However, here, our model could not predict the behavior of the line intensity, perhaps because the dependence of \( \sigma \) on \( f_{\text{ox}} \) is not the same as for Mo.

**C. Time dependence of intensity**

When a target is exposed to oxygen without the presence of the ion beam the oxygen coverage will increase to some equilibrium value; if now the oxygen is shut off and the ion beam returned to the target, there is a reduction of surface coverage due to sputtering. Under these conditions \( P_A \) becomes negligibly small and the solution to Eq. (3) is then given by

\[
f_{\text{ox}} = C \exp[-(\gamma' / \gamma)],
\]

where \( \gamma \) is equal to \( N_{\text{ox}} / I_B S_{\text{ox}} \) and \( C \) is a constant of integration. The time dependence of the photon emission intensity is obtained by substituting Eq. (9) into Eq. (2) which yields

\[
\Gamma = \gamma + \left[ C \exp[-(I_B S_{\text{ox}} / N_{\text{ox}})] \right] (\gamma' / \gamma).
\]

An exponential decay of the emission intensity was, in fact, observed when an ion beam was first directed onto a Mo target with oxygen already present. From this data at 20 keV, \( \tau \) was found to be \( 1.35 \times 10^7 \) sec which implies a value for \( S_{\text{ox}} \) of 0.0022 (the atomic surface density of molybdenum was calculated to be 2 \( \times \) 10\(^{15}\) atoms/cm\(^2\)).

**D. Dependence of intensity on beam current**

The integrated emission intensity as a function of the beam current \( I_B \) is given by \( I \) in Eq. (8). At very low oxygen pressures (\( \sim 10^{-11} \) Torr) the factor \( (2I_B S_{\text{ox}} kT) / PV \) is very much greater than unity. Therefore, the intensity \( I \) is dominated by the first term and is given by

\[
I = I_B \gamma.
\]

At very high oxygen pressures (\( \sim 10^{-5} \) Torr) the term \( 2I_B S_{\text{ox}} kT / PV \) is much less than unity. In this case \( I \) is given by

\[
I = I_B \gamma' + I_B \gamma'(\gamma' / \gamma) = \gamma' I_B.
\]

In both cases above \( I \) is linearly proportional to \( I_B \). At intermediate pressures, however, this is no longer true because \( 2I_B S_{\text{ox}} kT / PV \) is comparable with unity. Figure 6 shows the predictions and the data on beam current dependence at 10\(^{-6}\), 10\(^{-11}\), and 3 \( \times \) 10\(^{-9}\) Torr oxygen pressures.
pressure. In each case the predictions are consistent with the data.

E. Molecular effect

One might reasonably anticipate that an H₂ ion incident on the surface should dissociate into two hydrogenic particles which then proceed to enter the target and scatter from it in an uncorrelated manner. Thus an H₂ ion incident at energy E should behave like two incident protons of energy $\frac{1}{2}E$. In Fig. 7 we show the measured emission coefficient $\Gamma$ for 20-keV H₂ impact on Mo in the presence of oxygen at various pressures. The measured $\Gamma$ at low oxygen pressures is in fact twice that for H⁺ impact at the same velocity; this is as expected. However at higher pressures the value of $\Gamma$ measured with H₂ is dependent on beam current and only for low currents does it approach twice the measured value for H⁺ impact at $\frac{1}{2}E$. In Fig. 7 we show the predicted pressure dependence determined from Eq. (8) using the $Y$ and $\gamma'$ values appropriate to the experimental measurements at low beam currents; the sputtering coefficient $S_{ox}$ used in this prediction is 0.022 which is twice that determined experimentally from the time dependence of intensity during sputtering of the oxide surface. Clearly the predicted curves are not in agreement with the experiment.

In Fig. 2 we show the energy dependence of the measured $\Gamma$ for H₂ impact on Mo for both clean surfaces (i.e., oxygen partial pressure of $10^{-6}$ Torr) and a surface in the presence of O₂ at $10^{-6}$ Torr; the data are to be compared with corresponding figures for H⁺ impact on the same graph. We note here that at each energy the data for H₂ ions at high oxygen pressure was taken at the same beam current density (40 nA/cm²). At low oxygen pressures the $\Gamma$ for H₂ at energy $E$ is twice that for H⁺ at energy $\frac{1}{2}E$ for all energies studied. Indeed this same result was shown in our earlier paper. This result suggests that H₂ dissociates to two hydrogenic particles whose further scattering in the target is uncorrelated. For the data at higher oxygen pressures the measurements with H₂ at energy E are approximately twice that for H⁺ at energy $\frac{1}{2}E$. The similarity between H⁺ and H₂ energy dependences and the fact that they follow the predictions of Eq. (1) suggests that the deexcitation parameter $A_{21}$ is the same for both H⁺ and H₂ ($[1.25 \pm 0.05] \times 10^{-12}$ cm·sec). However for this high-pressure data the measured $\Gamma$ is slightly dependent on the beam current used for the measurements as shown in Fig. 7. Thus for the high-pressure data where oxygen is present on the surface the measured emission coefficient $\Gamma$ for H₂ is not exactly twice that for the corresponding situation for H⁺ impact; this we shall term the molecular effect.

The origin of the molecular effect is not clear. The deexcitation coefficient is (from Fig. 1) apparently not greatly different from the case for H⁺ impact. Thus the molecular effect represents either the influence of a new mechanism for excited hydrogen formation which is not present for H⁺ impact or else it implies that the probability for excited state formation $[F]$ of Eq. (1) is a function of the nature of the incoming projectile. A further possibility is that either the functional form for $O$ [Eq. (4)] or the whole model is too simplistic.

We can find no evidence for the presence of additional processes leading to excited hydrogen formation only in the case of H₂ impact. One might argue that excited hydrogen might be formed by sputtering of previously implanted H⁺ that has diffused to the surface and perhaps has formed a bond with oxygen on the surface. This can be ruled out since such sputtered hydrogen will be slow and would show up in the spectral line shape as a contribution at an essentially unshifted wavelength; this is not observed (see Fig. 1). Moreover this type of mechanism should be observed also for H⁺ impact. Also emission which is associated with the OH group itself (e.g., the $^2\Sigma^+ \rightarrow ^2\Pi$ transitions emitting between 302 and 309 nm) was not detected. In fact all of the emission in the spectrum (250–600 nm) was identified either as Balmer-H₂ or Mo I emission or as an underlying continuum which was reported previously.

It is known that molecular projectiles can be scat-
tered undissociated from surfaces; if these were excited they might dissociate to give excited atoms which will contribute at a large Doppler shift. Such scattered molecules should be observed only at small scattering angles and should decrease rapidly as incidence angle is increased. Experiments have shown that the discrepancy between \( \Gamma \) at low and high beam currents is independent of incidence angle; thus we conclude that scattered molecules are apparently not the origin of “the molecular effect”.

There remains only the possibility that the general assumptions used to formulate \( \gamma \) [Eq. (1)] and \( \Gamma \) [Eq. (8)] are defective. For example, the probability for formation of excited states, \( F_j \), has been assumed constant; this has been justified only through good agreement between experiment and theory for the \( H^+ \) impact data. Second, the assumptions concerning sputtering and sticking probability used to derive Eq. (8) are clearly simplistic and again are justified primarily by the success of the model in predicting the behavior for \( H^+ \) impact.

IV. CONCLUSION

For \( H^+ \) and \( H_2^+ \) ions incident on Mo and Cu we have shown that the emission intensity from backscattered \( H \) atoms changes with oxygen coverage. Our data on line shape analysis and energy dependence of the intensity for \( H^+ \) on Mo suggests that the intensity changes should be associated with a change in the \( A/a \) coefficient of Eq. (1), but not with changes in \( F_j \), the fraction of backscattered hydrogenic particles formed in the state \( j \). The \( A/a \) coefficient can be related to the probability that a nonradiative decay (probably the Auger effect) can occur for a backscattered excited \( H \) atom close to the target surface. Based on the changes in \( A/a \) with oxygen coverage we formulated a model which predicts the intensity as a function of beam current, projectile energy, and oxygen pressure, but only for \( H^+ \) ions incident on Mo. An attempt to extend the model to predict the photon emission from Mo bombarded by \( H_2^+ \) ions was not successful.

Appendix iii


Excited State Formed by B\(^+\) Impact on Metallic Cu and Mo.\(^+\)

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Abstract

Impact of 60 to 200 keV Be\(^+\) ions on polycrystalline Cu and Mo targets gives rise to emission from backscattered excited Be and Be\(^+\). The emission coefficients for these lines are presented as a function of impact energy and analyzed to provide a measure of the survival parameter, \(A/a\), for the atom or ion recoiling from the target. For B recoiling from both Cu and Mo this is found to be \(1.3 \times 10^8\) cm/sec while for recoiling B\(^+\) it is \(2.4 \times 10^8\). In the case of recoiling B\(^+\) the radiationless de-excitation must be by the Auger mechanism. Emission coefficients for the 3247, 3274 Å CuI doublet are also presented and rise slowly throughout the energy range studied.
I. Introduction

Impact of energetic ions on surfaces gives rise to light emission from sputtered and reflected particles as well as from excitation within the solid itself. We have performed extensive studies of 5 to 30 keV hydrogen and helium ion impact on metals where the principal source of emission is from backscattered excited atoms; the Doppler broadened spectral lines may be analyzed to provide information on the velocity distribution of the backscattered projectiles and on the radiationless de-excitation of the recoiling species. H\(^+\) and He\(^+\) impact at 5 to 30 keV energy do not induce significant emission from sputtered target particles. In certain cases continua characteristic of the metal target have been identified.\(^6\)

The present paper is concerned with the study of light emission induced by B\(^+\) impact on Cu and Mo. Impact energies are 60 to 200 keV providing an impact velocity range which is similar that employed in our earlier studies using incident H\(^+\) and He\(^+\). In common with the earlier work with incident H\(^+\) and He\(^+\) ions we observe Doppler broadened emission from backscattered, neutralized, excited projectiles. In addition there is copious emission from backscattered excited ions and weak emission from sputtered target atoms. Surveys of spectra induced by heavy rare gas ions (Ne\(^+\) and Ar\(^+\)) indicate that for these heavier species the emission is due primarily to sputtered neutral particles, and little emission from backscattered projectiles is seen. Thus the present work with B\(^+\) impact provides an unusual opportunity to examine simultaneously the behavior of primary ion scattering, primary ion neutralization and target sputtering.
II. Experimental Techniques

The experimental facilities and procedures remain essentially the same as described earlier\(^1\); therefore no detailed description will be provided here. A momentum analyzed beam from an accelerator is directed onto a metallic target and the light emission from the impact point is analyzed with a scanning monochromator. The target environment has a $2 \times 10^{-9}$ torr base pressure and is satisfactorily isolated from the accelerator by differential pumping. Optical observations are made through a sapphire window.

The angle of ion impact on the surface could be varied but was maintained at 60° (with respect to the surface normal) for all the work discussed here. The axis of optical observation was perpendicular to the ion beam and in the plane defined by the target normal and the incident ion beam. A spectral range of 2000 Å to 6000 Å was accessible to the optical system; no attempt was made in the present work to determine detection sensitivity.

The targets utilized here were high purity polycrystalline foils from the Materials Research Corporation; before use they were mechanically and electrically polished. The ion accelerator used in the present work was a 200 keV ion-implanter from Accelerators, Inc. fitted with a hot filament source. This replaces the 5 to 30 keV machine used in our earlier work\(^1-6\) and constitutes the only substantive equipment change.

The data presented here are generally the intensity of an atomic line integrated over its Doppler broadened width, presented as a function of incident ion energy. No calibration of detection sensitivity was performed so comparisons of line intensities at different wavelengths are only qualitative. Nevertheless for a specific $\text{B}$ or $\text{B}^+$ line the relative intensity for different targets is a valid comparison since of course the wavelength...
is the same. For a given spectral line the variation of relative intensity with projectile energy and with changes in target material are accurate to ±10%; this figure represents principally the reproducibility of the data.

The data are emission coefficients $\gamma_{ij}$ for a transition from a state $i$ to a state $j$ expressed in terms of the number of emitted photons per ion incident. In the event that the state $i$ is populated only by the basic surface collisions (and not appreciably by cascade from higher states) then the emission coefficient is proportional to the excitation coefficient $\gamma_i$ defined as the number of excited recoils formed per ion incident. The relationship is discussed in detail by Kerkdijk and Thomas. Influence of cascade population can be studied by monitoring directly the intensity of the cascade transitions. No cascade transitions are listed in the tables of Weise et al., for the excited states studied here so would conclude that such transitions are of low probability and therefore that we may take the measured emission coefficients $\gamma_{ij}$ as being proportional to the excitation coefficients $\gamma_i$.

Certain of the emissions (notably those from backscattered $B^+$ ions) were very sensitive to the deliberate introduction of oxygen into the vacuum environment; they also showed some variation of intensity with time after initial bombardment was commenced representing presumably the removal of a surface oxide film by sputtering. To overcome the influence of such contamination all samples were subject to initial sputter cleaning with a 100 keV $Ar^+$ ion beam from the accelerator, and the partial pressure of $O_2$ in the target chamber was maintained below $5 \times 10^{-11}$ torr.

There was concern that the implantation of $B^+$ ions into the target during the experiment, might cause changes to surface conditions and so influence the emission intensities. The presence of implanted boron was
confirmed by directing an \( \text{Ar}^+ \) beam onto Cu and Mo target that had been previously subject to a dose of \( 5 \times 10^{18} \ \text{B}^+ \) ions/cm\(^2\) and then observing the optical emission spectrum from the bombardment point. BI lines (2497 A and 2089 A) as well as BII lines (3451 A) were both observed indicating presence of boron, along with the expected emissions from sputtered substrate atoms. No evidence was found that the line intensities induced by \( \text{B}^+ \) impact on Cu and Mo were dependent upon the dose of boron ions to which the target had been subjected; we conclude therefore that the surface properties governing formation of excited B and \( \text{B}^+ \) recoils are not appreciably altered by the presence of implanted boron.

III. General Spectral Characteristics

Fig. 1 shows a typical spectrum observed for 1 keV \( \text{B}^+ \) impact on Cu. The lines at 2089 and 2497 A are identified as BI transitions (\( 2p^2 \ 2^2\text{D} \rightarrow 2p^2 \ 2^2\text{P}_0 \) and \( 3s \ 2^2\text{S} \rightarrow 2p \ 2^2\text{P}_0 \) respectively). The intense line at 3451 A is the \( 2p^2 \ 1^2\text{D} \rightarrow 2p \ 1^2\text{P}_0 \) transition in BII. Also observed weakly (but not within the spectral range of Fig. 1) is the 4121 A, \( 4f \ 3^3\text{P}_0 \rightarrow 3d \ 3^3\text{D} \) transition of BII. These various boron lines are observed with both Cu and Mo targets. From 3000 A to 3500 A for both Cu and Mo targets there is a broad underlying continuum. The origin of the continuum is not fully understood but may be related to fluorescence of the window through which optical observations were made. Superimposed on the continuum, at about 3250 A is a broad peak which is CuI doublet at 3248 A and 3274 A; at the resolution used in this scan (48 A) the doublet is unresolved. This CuI doublet occurs only for the Cu target. For Mo targets there are no significant emissions from...
molybdenum lines and no indication of the continuous spectra observed only when oxygen is present and tentatively identified as due to sputtered MoO.

Spectra have been recorded at a higher resolution (8 Å) to search for structure, shifts and broadening. We could detect no shift or broadening of the CuI lines at 3248 Å and 3274 Å which are due to sputtered copper; according to Van der Weg there should be shifts and broadening of about 1 Å on these lines representing the energy distribution of sputtered particles but such small effects would not be disclosed under the resolution employed here (8 Å). The BI and BII lines are both broadened with the peak intensity shifted to the blue; the shape is similar to that from backscattered H* and He*. The shift and line width varies with incident projectile energy and is consistent with the backscattering model invoked previously by ourselves and others for lighter projectiles. Thus the shape and shift are consistent with the source of emission being backscattered projectiles.

We studied the effect of introducing oxygen into the chamber to raise the total pressure from the normal base value of 2 x 10^{-9} torr to 6 x 10^{-8} torr. For 100 keV B⁺ on both Cu and Mo the intensity of the BII lines increased by a factor of three while the intensity of the BI lines changed by only 10%. For the Mo target, with oxygen present, we observed also the appearance of the broad band spectrum seen previously with Ar⁺ ion impact and ascribed by us to sputtered MoO; the most distinct feature was the characteristic narrow band observed at 2960 Å.
 indication of the continuous spectrum observed only oxygen is present and tentatively identified as due to sputtered MoO.

Spectra have been recorded at a higher resolution (8 Å) to search for structure, shifts and broadening. We could detect no shift or broadening of the Co I lines at 3248 Å and 3274 Å which are due to sputtered copper; according to Van der Weg \(^\text{10}\) there should be shifts and broadening of about 1 Å on these lines representing the energy distribution of sputtered particles but such small effects would not be discerned even at the resolution employed here (8 Å). The Bi I and Bi II lines are both broadened with the peak intensity shifted to the blue; the shape is similar to that of backscattered H\(^+\) and He\(^+\) \(^\text{2}\). The shift and line width varies with incident projectile energy and is consistent with the backscattering model invoked previously by ourselves \(^\text{1-5}\) and others \(^\text{11}\) for lighter projectiles. Thus the shape and shift are consistent with the source of emission being backscattered projectiles.

We studied the effect of introducing oxygen into the chamber to raise the total pressure from the normal base value of 2 \(\times\) 10^-8 to 1.6 \(\times\) 10^-8 torr. For 100 keV H\(^+\) on both Cu and Mo the intensity of the Bi II lines increased by a factor of three while the intensity of the Bi I lines changed by only 10%. For the Mo target, with oxygen present, we observed also the appearance of the broad band spectrum seen previously with Ar\(^+\) ion impact and described by us \(^\text{9}\) to sputtered MoO; the most distinct feature was the characteristic narrow band observed at 9 2900 Å.
IV. Emission from Backscattered Boron

Figs. 2 & 3 show the dependence on incident B⁺ projectile energy of emissions from backscattered B atoms and B⁺ ions; included are data for the 2497 Å BI and the 3451 Å BII line. Measurements on the 2089 Å BI line and the 4122 Å BII lines (not shown here) showed the same relative dependence on projectile energy as the 2497 Å BI and 3451 Å BII lines respectively. Clearly the relative energy dependence for a given line is the same for both Cu and Mo targets although intensity is larger in the latter case.

In our previous studies of backscattered H⁺ and He⁺ from metals we have developed theoretical descriptions which successfully model the intensity of emission from backscattered particles. McCracken and Freeman¹² provide a prescription to predict the flux of projectiles emerging from the surface with a velocity component normal to the surface of between \( v_\perp \) and \( v_\perp + dv_\perp \); we shall write this fraction as \( f(v_\perp, E_0, \phi) \), where \( E_0 \) is the original energy at impact and \( \phi \) is the angle of incidence. Of these backscattered particles a certain fraction, \( F_i \), are in the excited state \( i \). Following our previous work¹² we shall assume this fraction is independent of both emergent angle and emergent energy. Finally we must allow for the possibility that some of the recoiling exited atom suffer radiationless de-excitation by Auger or resonant transitions while within a few Angstroms of the surface; this is accommodated by a factor \( R(v_\perp) = \exp(-A/av_\perp) \) representing the probability of scattered particle with perpendicular velocity, \( v_\perp \), escaping without radiationless de-excitation. The two parameters \( A \) and \( a \) are dependent on the potential barrier, the wave functions of the participating electrons and on the nature of the de-excitation mechanism. In this manner one may formulate the excitation coefficient \( \gamma_i(E_0, \phi) \) for formation of excited recoil atoms in the state \( i \) when projectiles of energy \( E_0 \)
are incident at an angle $\phi$ to the surface normal:

$$\gamma_i(E_0,\phi) = \int_{v_\perp}^{u} f(v_\perp,E_0,\phi) F_i \exp\left(-\frac{\Lambda}{av_{\perp}}\right) \, dv_{\perp} \tag{1}$$

The upper limit of the velocity integral corresponds to scattering from an atom on the surface; to establish a lower limit of integration we assume that any projectile which has its energy reduced to 20 eV will be trapped in the lattice and will not escape. Since there is no reliable estimate of the ratio $A/a$ we perform the computation for a range of values to search for an agreement between experiment and the predictions of Eq.(1). No attempt is made to predict absolute values of $\gamma_i$ and the computed values are compared only with the relative measured values of emission coefficients as a function of incident projectile energy.

Fig. 2 shows the measured emission coefficient of the 2497 Å (3s $^2$S $\rightarrow$ 2p $^2$P) BI transition along with predictions of the excitation coefficient obtained from Eq.(1); experiment and model calculations are normalized together at 60 keV. The model calculations using $A/a = 1.3 \times 10^8$ cm/sec provide a good agreement with experiment; the other computations shown with survival coefficients of $1.5 \times 10^8$ cm/sec and $1.1 \times 10^8$ cm/sec agree less satisfactorily with experiment. We conclude that the model agrees with the experimental data and that a survival coefficient of $1.3 \times 10^8$ cm/sec is appropriate; the precision with which the survival coefficient is established is about 12.5%.

Fig. 3 shows the measured emission coefficient of the 3451 Å (2p$^2$ $^1$D $\rightarrow$ 2p $^1$P$^0$) BI line along with certain predictions of excitation coefficient from Eq.1; experiment and computation are normalized at 150 keV for Mo and 160 keV for Cu, clearly none of the predicted curves provide a
close representation of the experimental data. We consider that use of a survival coefficient $A/a$ of $2.4 \times 10^8$ cm/sec in the model provides a qualitative agreement with the general form of the data; survival coefficients of $3.0 \times 10^8$ or $2.0 \times 10^8$ are less satisfactory. It is not clear why the model computations in this case do not satisfactorily represent the experimental data. It is to be noted that the model provides a precise representation of emission coefficient for recoils which are neutral as for recoiling B in the present work and recoiling H and He in our earlier studies; this is the first case where the model has been applied to a recoiling ion and here it appears to be inadequate. The origin of the discrepancy may well lie in the assumption that $F_1$ (the probability of a recoil being in the relevant excited state) is constant; there is no independent justification for this assumption.

In our earlier studies of $H^*$ and $He^*$ backscattering we have shown that the appropriate survival coefficients $A/a$ are in the range $7 \times 10^7$ cm/sec to $3 \times 10^8$ cm/sec depending somewhat on the excited level under study. Thus the present work is governed by survival coefficients of very similar magnitude. It is worth noting that the present work using Boron involves essentially the same incident projectile velocities as the earlier work with hydrogen and helium. There are no reliable theoretical estimates of the coefficient $A/a$ but approximate calculations by Cobas and Lamb as interpreted by Hagstrum predict a value of $2 \times 10^8$ cm/sec for an Auger de-excitation of metastable helium on molybdenum; this is fairly close to the values derived from the present work.

Determination of the survival parameter $A/a$ by this method provides no information on whether the mechanism is a resonant transition or an Auger decay. Hagstrum provides a simple distinction between the two mechanisms based on the energy levels of the participating electrons. Let $E_i$ be the energy required to ionize the recoiling species if in its ground state and $E_e$ be the
excitation energy of the excited state of interest; then $E_i - E_e$ is the binding energy of the excited electron. This energy may be compared with the work function $\phi$ of the metal from which the projectile is recoiling. In the Auger mechanism an electron from the metal decays to the ground state of the recoiling species giving its energy to the excited electron and ejecting it to the continuum.

The resonant decay occurs by tunneling of the excited electron through the potential barrier into a vacant state above the Fermi level; it can occur only if $E_i - E_e > \phi$. All of our previous work $^{1,2,3}$ and that of others $^{11}$ on backscattering of excited H and He has involved excited states where $E_i - E_e > \phi$; from an energetic point of view both Auger and resonance decay modes are possible.

By contrast, for the case of the 3451 Å BII line the relevant excited state ($2p^2 1D$) has an energy of 9.10 eV and the ionization potential of $B^+$ is 25.15 eV giving $E_i - E_e$ a value of 16.05 eV which clearly exceeds the work function of the metals used here. So the $2p^2 1D$ state can not undergo radiationless decay by the resonant ionization mechanism; and the A/a value determined for this state must be for the Auger mechanism. We note that the 4121 Å line from the $3p^0$ BII state has the same energy dependance implying that the $3p^0$ state also decays by the Auger mechanism.

The increase in emission coefficient observed when oxygen is introduced into the target chamber is consistent with a reduction in the A/a value, permitting a greater fraction of the excited recoils to escape without de-excitation. This behavior has been previously observed $^{15}$ in the study of excited hydrogen backscattering from surfaces.

V. The CuI line of Sputtered Copper

In fig. 4 we show the intensity of the CuI doublet lines 3247 Å and 3274 Å, measured together, as a function of incident $B^+$ energy. Clearly
the emission coefficient rises slowly through the energy range of the present experiment with no significant features.

In earlier studies of sputtered excited atoms performed with low (30 to 3000 eV) projectile energies White and Tolk\textsuperscript{16} have attempted to model the emission coefficient using a formulation similar to our Eq. 1; they include the possibility of radiationless decay mechanisms and estimate a value for the $A/a$ parameter. In performing the same analysis for the present work one is faced with the difficulty of estimating the factor $f(v / E_0, \phi)$ which would now represent the velocity distribution of sputtered particles induced by impact of a projectile of energy $E_0$ at an angle $\phi$. According to the Sigmund theory of sputtering\textsuperscript{17} the energy distribution of sputtered particles is inversely proportional to the square of ejection energy; moreover the total sputtering coefficient is proportional to the nuclear stopping power. We have used such formulations to predict the emission coefficient employing the $A/a$ value of $2 \times 10^6$ cm/sec determined previously by White and Tolk.\textsuperscript{16} It is found that the predicted energy dependence is governed primarily by the nuclear stopping power; according to LSS theory\textsuperscript{18} this peaks at about 6 keV for B$^+$ impact and declines to higher energy. Clearly this in no way represents the present data where the emission increases with energy to 200 keV; this leads to the possibility that the basic excitation probability ($F_i$ of Eq. 1) is dependent on projectile energy. Such a dependence was in fact found by White and Tolk\textsuperscript{16} in their study of excited Cu formation in low energy sputtering. In the absence of reliable information on the factors required to predict $F_i$ for sputtering we do not attempt to model the data shown in Fig. 3.

\textbf{VII. Conclusion}

Impact of B$^+$ ions on Mo and Cu produces BI and BIT emissions from backscattered neutral atoms and ions. The dependence of intensity on projectile energy has been modeled and is governed primarily by the radiationless decay of the recoiling...
excited state. Survival coefficients have been determined and are of similar magnitude to those for backscattering of excited B and He found previously. The parameter $A/a$ is of the order $1.3 \times 10^8$ cm/sec for recoiling $B^*$ and $2.4 \times 10^8$ cm/sec for recoiling $B^{**}$. In the case of $B^{**}$ only the Auger de-excitation mechanism is energetically possible.
† Supported in part by the Magnetic Fusion Energy Division of the Energy Research and Development Administration.

References


Figure Captions

1. Spectrum induced by 180 keV B^+ impact on Cu. The underlying continuum from 3000 Å to 3500 Å is due to fluorescence of the window through which observations are made.

2. Relative emission coefficient for the 3s \(^2S\) \rightarrow 2p \(^2P_1\) BI transition induced by B^+ impact on Cu (squares) and Mo (circles). The full lines are the predictions of the model using A/a = 1.3 \times 10^8 cm/sec, normalized to experiment at 60 keV impact energy. The dashed lines show the predictions of the model for (a) A/a = 1.5 \times 10^8 cm/sec and (b) A/a = 1.1 \times 10^8 cm/sec for the case of B^+ on Mo only.

3. Relative emission coefficient for the 2p \(^2P_0\) \rightarrow 2p \(^2P_1\) transition induced by B^+ impact on Cu (squares) and Mo (circles). The solid lines are drawn only to indicate the trend of the data. The dashed lines represent predictions of the model; (a) A/a = 2.4 \times 10^8 cm/sec shown both for Mo and for Cu, (b) A/a = 3.0 \times 10^8 cm/sec, shown only for Cu (c) A/a = 2.0 \times 10^8 cm/sec, shown only for Cu.

4. Relative emission coefficient for the 4 \(^2P_0\) \rightarrow 4 \(^2S\) CuI doublet induced by B^+ impact on Cu. The line is drawn only to indicate the general trend of the data.
Fig 1
Fig 2
INCIDENT ENERGY (keV)

EMISSION COEFFICIENT $\gamma_i$ (ARBITRARY UNITS)

- (A)
- (B)
- (C)

Mo
Cu
I. **OBJECTIVE**

A small fraction of sputtered atoms are ejected in excited states; emission from their radiative decay may be detected and used to provide at least qualitative information on the composition of the surface. This technique of surface analysis is called SCANIIR (Surface Composition Analysis by Neutral and Ion Induced Radiation). In many respects this approach is similar to SIMS (Secondary Ion Mass Spectrometry) where one observes the mass spectrum of sputtered particles. In both SCANIIR and SIMS it is possible to detect signals from ejected molecules and achieve some indication of surface chemistry.

The objective was to assess the potential usefulness of SCANIIR in detection of H, C, O, and metals normally found in fusion device plasmas. The experimental set up is the same as that used by us for studies of excited state backscattering and it has been well described in the literature\(^1\). A heavy ion beam (e.g. 10 to 200 keV Ar\(^+\)) is directed onto the surface and the emission detected with a scanning monochromator. The work described here utilized a monochromator spanning the range 2000 to 8000 Å; We are currently performing additional studies in the range 900 to 2000 Å. We can study clean surfaces to establish the spectrum of the substrate and the wavelengths at which a given species emits most intensely. Then we must provide a "contaminated" substrate to establish whether small quantities of material can be detected in the presence of a different substrate.

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Such contaminated surfaces can be produced by either implanting ions to a shallow depth or by adsorbing a gas onto a surface.

II. GENERAL SPECTROSCOPIC OBSERVATIONS

It has become clear that intense signals are obtained only from transitions to the ground state of the ejected species; transitions between excited states tend to be very weak. Thus we have detected many metals but have failed to readily detect H, C, and O for which the transitions to ground state occur in the vacuum u-v spectral region; in the case of C we have detected weak emission at visible wavelengths but not at useful intensities. In Table II we list some of the species studied and the wavelengths which are most convenient for detection. We indicate qualitatively the intensity of emission. A "weak" designation indicates that the emission intensity is comparable with background continua and detector noise; it will be impossible to use such a "weak" line for analytical purposes.

<table>
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</tr>
<tr>
<td>B</td>
<td>BI</td>
<td>Strong</td>
<td>2089</td>
</tr>
<tr>
<td>C</td>
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<td>3737†</td>
</tr>
<tr>
<td>Cu</td>
<td>CuI</td>
<td>Strong</td>
<td>3247</td>
</tr>
<tr>
<td>Mo</td>
<td>MoI</td>
<td>Strong</td>
<td>3798†</td>
</tr>
</tbody>
</table>

†Numerous other useful lines also observed.
We are currently setting up to search for the H, C and O emissions in the vacuum u-v and anticipate having performed preliminary experiments within one month of this report's date. Scharmann (University of Giessen) has informed us privately that he has detected these species from uncontrolled heavily contaminated targets and we must confirm whether they are seen from targets with small controlled contaminant levels. In general terms we conclude that metals can readily be detected using transitions to the ground state and we anticipate that H, C and O can also be detected by the corresponding transitions which lie at vacuum u-v wavelengths.

III. PROFILING EFFECTS

The SCANIIR technique is in a sense destructive at the microscopic level since it removes the surface layer to expose the underlying material. This does introduce certain minor complications. Certainly the surface composition will change and possibly the surface contaminants will be driven into the target by knock-on collisions or radiation enhanced diffusion. We shall discuss two examples of these.

In Fig. 1 we show the intensity of CrI and FeI lines when type 304 Stainless Steel is bombarded with Ar⁺. It is observed that the Cr emission drops rapidly as the surface is eroded. This represents the well known fact that chromium segregates to give a higher density at the surface than in the bulk. Modest temperature cycling (to 300°C) did not cause the Cr signal to return to its original level.
Fig. 1. Time dependence of optical signals from Cr and Fe when profiling a type 304 Stainless Steel sample using a 100 keV Ar$^+$ beam.
In Fig. 2. we show emissions from a Cu line resulting from Cu implanted into carbon to a dose of $1.44 \times 10^{15}$ atoms in about $10 \text{ mm}^2$ with an energy of 100 keV;

![Graph](image)

Fig. 2. Time dependence of CuI line (3247 Å) when profiling a carbon sample with implanted Cu. The profiling beam is 100 keV Ar$^+$. The emission is induced by 100 keV Ar$^+$ impact on this implanted target. One would have anticipated that the Cu emission should have peaked and then returned to zero providing a depth profile of the implant. Indeed a maximum is observed (at a profiling dose of about $2 \times 10^{15}$ atoms) but the intensity does not decrease to zero and remains high for an indefinite period. Copper is of course insoluble in carbon and will therefore tend to migrate to damage sites. We believe that the
profiling Ar$^+$ beam is producing damage beyond the implant giving rise to radiation enhanced diffusion into the bulk. This can of course be eliminated in an analytical situation by utilizing a low energy Ar$^+$ beam to minimise the depth of damage. We are currently analysing these profiling data to determine the effective diffusion constants.

IV. DETECTION SENSITIVITY

We are currently evaluating detection sensitivities by comparing optical signals with contaminant density determined by Auger spectroscopy and Rutherford backscattering. These are not yet complete due late arrival of the required surface analysis facilities. However some estimates can readily be made.

From the data of Fig. 2, we have a integrated signal from Cu, to the point where the run was terminated, of some $10^5$ photons, resulting from an implanted contaminant of approximately $10^{15}$ Cu atoms in a 10 mm$^2$ area. A reliable signal should be no less than background noise; such noise is of the order $10^4$ counts for the period shown in Fig. 2. Thus we would argue that a copper implant of $10^{14}$ atoms in a 10 mm$^2$ area can be reliably detected; this is one monolayer. Detection efficiency can be increased by three or more orders of magnitude by utilizing a photomultiplier-filter combination tuned to the wavelength of interest; such single channel arrangement would be ideal for following the time history of a specific species in a MFE device. Thus a detection limit of $10^{12}$ atoms/cm$^2$ is achievable ($10^{11}$ atoms on 10 mm$^2$). Similar figures are achieved for Cr, Fe, Mo etc. We note that recent studies$^2$ of the British DITE Tokomak device have shown metals to be deposited on the walls at a concentration of $10^{15}$

atoms / cm$^2$ in one discharge. Thus we would argue that the SCANIIR system can
certainly detect the deposition of a single discharge and has the potential
for time resolution to intervals of $1/100$ of the discharge period.

The above conclusions arise from our study of metals. Clearly no fusion
device will operate with appreciable heavy metal impurity levels and in the long
term we should expect them to be unimportant; indeed these high levels of metals
may be peculiar to DITE. To be useful, the SCANIIR technique must also detect
the time history of H, C and O. As we noted earlier we anticipate that spectro-
scopy at 900 to 2000 Å will permit these species to be detected using transitions
to the ground states; we see no immediate reason why detection sensitivity should
not be similar to that for metals.

V. INTERIM CONCLUSION

We conclude that a single channel spectroscopic system (i.e. a filter-
photomultiplier combination) can be utilized to follow the time history of metal
deposition during the single pulse of a Tokomak machine. We have good reason
to believe (and are currently attempting to confirm) that this conclusion holds
also for H, C, and O deposition which will be the principle contaminants in
any fusion device.
Appendix v

Backscattering of 1 to 30 keV Hydrogen from Metal Surfaces

(A preliminary report by J. E. Harriss)

Abstract

A study has been made of the scattering of H\(^0\), H\(^+\), and H\(^-\) from metal surfaces during bombardment by energetic protons. Absolute measurement of scattered ion current is reported for 15 keV protons on copper at near-glancing angle. Energy spectra of these scattered ions peak around 9.5 keV for H\(^+\) and 8.5 for H\(^-\). Here the charged fraction of the scattered flux is about 25%, the ion flux being 87% H\(^+\) and 13% H\(^-\). The energy distribution predicted from McCracken and Freeman's theory is shown to be invalid for scattering about 10\(^\circ\) from the surface.
I. INTRODUCTION

When a beam of protons strikes a metal surface, many of the incident particles are scattered out of the metal. In general, because of the scattering process, these particles suffer changes in their direction, energy, and charge-state. Reported in the present work are results of measurements of all these changes for protons incident on polycrystalline copper. A number of theories exist which attempt to treat this interaction, and one goal of our work is to test the applicability of the theory by McCracken and Freeman to the prediction of angular and energy distributions.

II. THEORY

Details of McCracken and Freeman's theory have been reported elsewhere, and it will be treated only summarily here. Basically, the theory is a single-collision prediction for the energy and angular distributions of energetic particles scattered from an amorphous solid. Account is made of electron stopping of the projectile, and the Rutherford cross section is presumed to govern the atom-atom collision.

The geometry of the scattering event is shown in Figure 1. The projectile penetrates the target at an angle $\phi$ with respect to the surface normal and exits the target at an angle $\theta$ with respect to the initial beam direction.

Figure 1. Collision geometry. $\hat{n}$ is the surface normal.
III. EXPERIMENTAL MEASUREMENTS

The experimental objectives are to measure absolutely the angular, the charge-state, and the energy distributions of the $\text{H}^+$, $\text{H}^-$ and $\text{H}^0$ flux scattered from the surface. These data are then to be compared to the predictions of the theory of McCracken and Freeman.

The experimental apparatus has already been described, and only a brief description will be given here. Ions are produced in an rf-discharge ion source, accelerated, mass analyzed, and focused into the collision chamber. Steering plates then direct the beam through two collimating apertures and into a Faraday cup. The cup can be moved, allowing the beam to strike the target. The target manipulator is located at the hub of a detection arm which is rotatable up to 45° from the initial beam direction. On the arm are two collimating apertures to define the angle, a pair of deflection plates to separate the ions, and three Faraday cups to measure the scattered flux. For measurement of the energy distribution, the deflection plates on the rotating arm were replaced by a 90° electrostatic energy analyzer. The angle of acceptance of the detectors is 0.5 degrees, and the resolution ($\Delta E/E$) of the energy analyzer is estimated to be $\pm 0.02$.

The results reported here are only samples of our work for protons on a technical, polycrystalline copper target. The angle of incidence for this work was 70° with respect to the target-surface normal.

We have studied the variations in the number of particles scattered out of the target as the target surface is scanned through the beam path. Such surface profiles indicate that mechanically polished copper surfaces typically show a 7% variation in total scattered yield over the surface. As discussed previously,
electropolished samples show greater variations (20%). When a target is baked above 350°, its scattered flux is increased about 3%,\(^2\) and targets which have suffered radiation damage show a decreased yield from the damaged site\(^2\) (-6%, typically). It is apparent that the accuracy with which absolute measurements of scattering can be made is greatly restricted.

For particular angles \(\theta\) we have measured absolutely the ion current scattered from a mechanically polished target for incident protons with an energy of 15.00 keV. On the same beam spot these values have been reproducible to better than 3%. When this is coupled with other sources of error, we estimate the error in the final values to be \(+15\%\). For \(\theta = 30.4^\circ\) this ion current is determined to be

\[
\frac{(i_{+} + i_{-})}{I_{B} \delta\omega} = 0.024 \quad \text{(amperes out)}
\]

\[
\times \frac{\text{(amperes in)(steradian)}}{I_{B} \delta\omega}
\]

where

- \(i_{+}\) = scattered proton current
- \(i_{-}\) = scattered \(\text{H}^-\) current
- \(I_{B}\) = incident beam current
- \(\delta\omega\) = solid angle of detectors

An electrostatic energy analyzer was installed to monitor the scattered flux, and energy scans were made of both the \(\text{H}^+\) and the \(\text{H}^-\) flux. The results of these scans for \(\theta = 30.4^\circ\) are shown in Figure 2. Discrete integration (via Simpson’s rule) of the \(\text{H}^+\) and \(\text{H}^-\) curves reveals that at this angle the ion flux is 87% protons and 13% \(\text{H}^-\).
Combining these charge-state fractions with the measured ion current scattered at the same angle, we obtain an absolute measure of the proton and H\(^-\) currents. At \(\theta = 30.4^\circ\)

\[
\frac{i_+}{I_B \delta \omega} = 0.028 \quad \frac{(\text{particles out})}{(\text{particles in})(\text{steradian})}
\]

\[
\frac{i_-}{I_B \delta \omega} = 0.0041 \quad \frac{(\text{particles out})}{(\text{particles in})(\text{steradian})}
\]

Figure 2 also shows (normalized to experiment) the prediction by McCracken and Freeman's theory for our experimental configuration. The prediction seems to be an invalid one for the conditions of our work. The prediction, however, does not differentiate between charge-states, and we have not yet measured the energy distribution of the neutrals. The similarity of the shapes of the H\(^+\) and H\(^-\) curves does not cause us to expect the H\(^0\) distribution to be significantly different in shape.

Previously we reported\(^2\) an energy distribution measured experimentally by Meischner and Verbeek.\(^3\) This distribution is similar to the above prediction for the energy distribution except that the experimental curve shows a peak around 1 keV, with the flux decreasing toward zero at lower energies. Meischner and Verbeek's work is for a fixed scattering angle \(\theta\) of 135\(^\circ\); the incident angle \(\phi\) varies from 0\(^\circ\) (normal incidence) to 45\(^\circ\). These conditions are radically different from our own, and there is no reason to expect the distribution to be similar. One intuitively expects that particles scattered at large angles will have lower average energy than particles scattered at small angles. This
Figure 2. Energy distribution of scattered ions at $\theta = 30.4^\circ$ for 15 keV protons striking copper at 70° from normal. Dots (●) are experimental data for scattered protons; triangles (△) are experimental data for scattered $H^-$; solid line is theoretical prediction based on McCracken and Freeman's theory and normalized to $H^+$ experimental curve at 5 keV.
expectation is consistent with the differences between our experimental energy
distribution and that of Meischner and Verbeek.

There is other evidence in support of our measurements. Morita et al. have measured angular and energy distributions for protons on vacuum-deposited copper with angular orientations similar to those in our experiments. For a 15.2 keV beam incident on the target at 80°, they detect an energy distribution quite like our own. Their published curve bears little detail especially at the energy extremes, however.

The typical angular distribution for 15 keV protons on copper at φ = 70° has already been reported. Further studies have verified the shape of the distribution, rising steeply from zero at the surface (θ=20°) to a maximum at 13° above the surface (i.e., at θ = 33°) and decreasing gradually at larger angles. Variations of ± 1° have been observed in the location of the peak, but values of scattered flux for angles in this range are separated by less than 2%.

The detector used for these measurements is a Faraday cup consisting of two plates (direct and scavenger) which may be monitored separately or as a single unit. When the plates are monitored separately, one obtains information about the scattered neutrals by studying the secondary emission of electrons from the direct plate. Careful analysis of this data indicates that for a 15.0 keV proton beam, 75% +3% of the flux scattered at θ = 30.4° is a neutral hydrogen.

IV. CONCLUSION

It is clear that variations in the scattering ability of a surface are significant and depend strongly on target history. However, careful studies of a particular beam spot yield measurements which are reproducible within several per cent. Such studies indicate that the scattered flux at θ = 30.4° is about 75% H°, 22% H⁺, and 3% H⁻ for 15 keV protons on copper at φ = 70°. Furthermore,
the scattered ion flux has been measured absolutely for the above situation.

The theory of McCracken and Freeman predicts reasonably well the general shape of the angular distribution of the scattered flux. However, for the relatively small scattering angles reported on here, the theory's prediction of the energy distribution of the scattered flux bears little resemblance to the measured distribution.

These sample data indicate in general terms the behaviour of our data. We are currently extending our data to other energies, and proceeding with similar measurements on Be, Mo and Stainless Steel.

REFERENCES


2. J. E. Harriss, Appendix v, "Backscattering of 5-25 keV Hydrogen from Metal Surfaces" in Formation of Excited Hydorgen Atoms by Charge Transfer and Dissociation, (E. W. Thomas, E. O. Rausch, J. E. Harriss, J. T. Bell, ORO-2591-79 (1976)).


**Appendix vi**

**List of Publications**

<table>
<thead>
<tr>
<th>Title</th>
<th>Authors</th>
<th>Journal/Status</th>
<th>Percentage ERDA Support</th>
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<tr>
<td>(2) &quot;Scattering of Protons from Surfaces, Charge State and Excited State Composition&quot;</td>
<td>E.W. Thomas, M. Zivitz, W.E. Baird, J.E. Harriss, E.O. Rausch</td>
<td>J. Nucl. Mat. 63, 205 (1976)</td>
<td>80%</td>
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<td>(3) &quot;Formation of Excited H by Impact of 5 to 30 keV H⁺, H₂⁺ and H₃⁺ Ions on Metal Surfaces.&quot;</td>
<td>E.O. Rausch, E.W. Thomas</td>
<td>Phys. Rev. A. 14, 1912 (1976)</td>
<td>80%</td>
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<td>(4) &quot;The Influence of Oxygen on Backscattering of Excited H when H⁺, H₂⁺, and H₃⁺ Ions of 4 to 30 keV Energy are Incident on Molybdenum&quot;</td>
<td>E.O. Rausch, M.W. Murray, H. Inouye, E.W. Thomas</td>
<td>J. Appl. Phys. 48, 4347 (1977)</td>
<td>80%</td>
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<td>(5) &quot;Excited H Formation by 2-200 keV H₁⁺, H₂⁺ and H₃⁺ Ion Impact on Metal Surfaces&quot;</td>
<td>E.O. Rausch, H. Inouye, A.J. Senol, E.W. Thomas</td>
<td>Phys. Rev. A (Accepted for Publication)</td>
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<td>(6) &quot;Excited State Formation by B⁺ Impact on Metallic Cu and Mo.&quot;</td>
<td>E.W. Thomas, H. Inouye, E.O. Rausch</td>
<td>J. Appl. Phys. (Accepted for Publication)</td>
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<td>(7) &quot;Backscattering of Excited Hydrogen from Mo in the Presence of Oxygen&quot;</td>
<td>E.O. Rausch, E.W. Thomas</td>
<td>J. Nucl. Instr. Meth. (Accepted for Publication)</td>
<td>80%</td>
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<td>(8) &quot;Formation of Excited H by Impact of H⁺, H₂⁺ and H₃⁺ Ions on Metal Surfaces&quot;</td>
<td>E.O. Rausch, E.W. Thomas</td>
<td>Proc. 7th Int. Conf. on Atomic Colls. in Solids (Moscow. U. Pub. House, in course publication)</td>
<td>80%</td>
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+ This publication is an abstract of an invited talk.