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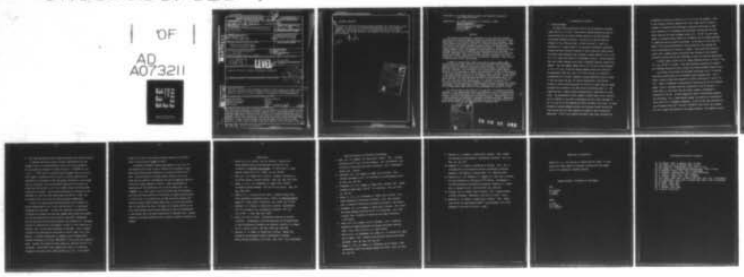

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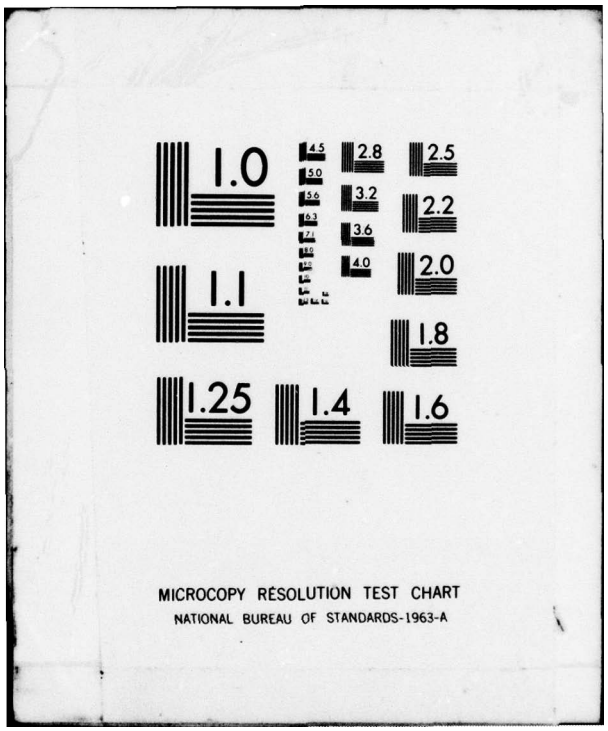
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The interaction mechanisms of selected ions and atoms with diatomic molecules were studied by making differential energy loss and time of flight measurements on the scattered beam under single collision conditions. Continued attention was given to the $He^+ + H_2$ collision system and it was shown that inelastic and charge exchange processes result from interactions occuring near particular projectile-target separations. A number of spectroscopic studies were made in which visible light, vacuum ultraviolet light, and soft x-rays were observed with sufficiently high

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20. ABSTRACT CONTINUED

resolution to identify the excited states produced. Ar-L and C-K soft x-rays from Ar^+ + Ar and Ar^+ + C collisions have been observed. A new computer-controlled, high-resolution soft x-ray spectroscopy facility has been established and used in studies of ion-induced x-rays both from gases and from solid surfaces.

Final Report: Low Energy Inelastic Atomic and Molecular Collisions - Grant DAAG-9-76-G-0130

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18 July 1979

Abstract

The interaction mechanisms of selected ions and atoms with diatomic molecules were studied by making differential energy loss and time of flight measurements on the scattered beam under single collision conditions. Continued attention was given to the $He^+ + H_2$ collision system and it was shown that inelastic and charge exchange processes result from interactions occuring near particular projectile-target separations. Studies of the charge exchange in $He^+ + H_2$ suggested a useful model for ion-molecule charge exchange, which correctly predicted the participating states and their relative importance in $Ar^+ + CO$ exchange collisions (as well as in the other systems we studied). This laboratory also verified a scaling law for the energy loss in electronically elastic ion-molecule collisions. Most recently a detailed study of $D^+ + H_2$ was begun.

A number of spectroscopic studies were made in which visible light, vacuum ultraviolet light, and soft x rays were observed with sufficiently high resolution to identify the excited states produced. The total optical emission cross sections were measured for several visible transitions in $He^+ + He$ collisions. For the helium P and D states with nonzero angular momentum, it was necessary to measure the polarization as well as the intensity of the light emitted at 90^o to the ion beam in order to account for the effects of collision-induced anisotropy on the emission cross sections. Vacuum ultraviolet emission cross sections were measured in the wavelength range of 40 - 110 nm for $Ar^+ + Ar$, $Ne^+ + Ne$, $Ne^+ + Ar$, $Ar^+ + Ne$, $He^+ + Ar$, $Ar^+ + He$, and $He^+ + Xe$ collisions at keV beam energies. In the $He^+ + Ar$ and $Ne^+ + Ar$ collisions, the cross section for producing the 92 and 93.2 nm lines of ionized argon peaks in the energy range studied. Conditions appear somewhat favorable for ion-beam pumped laser action at these wavelengths in the $He^+ + Ar$ case.

Ar-L and C-K soft x rays from $Ar^+ + Ar$ and $Ar^+ + C$ collisions have been observed. A new computer-controlled, high-resolution soft x-ray spectroscopy facility has been established in our laboratory; it is used in studies of ion-induced x rays both from gases and from solid surfaces. Characteristic Ar-L x rays from argon ions implanted in graphite have been observed. Experiments are planned to assess the general usefulness of soft x-ray spectroscopy as a probe of both depth and site distributions of implanted ions.

I. Ion-Molecule Collisions

A. Charge Exchange

The small angle scattering of He^+ by H_2 was studied in an energy range from 0.5 to 3.0 keV. Electrostatic energy analysis was used on the scattered He^+ and time-of-flight analysis on the He^o resulting from charge exchange collisions. The spectra of the He^+ show the presence of four dominant peaks, labeled A,B,C and D. Peak A is attributed to a "quasi elastic" (no electronic excitation) scattering process with both collision partners in their electronic ground states. The lowest lying inelastic peak, labeled B, always appears at an energy of 13.3 eV below that found for the maximum in peak A. The He^o spectra show that charge exchange occurs with a threshold inelastic energy loss, $Q \approx 13$eV. Over the angular range from 0.1 to 3.0^o and at beam energies of 0.5, 0.75, and 1.0 keV the charge exchange occurs with $13 < Q < 18$ eV corresponding to He^o (1s, $n \geq 2$) + H^+ + H(1s) as well as to $He^o(1s^2)$ accompanied by H^+ + H($n \geq 1$) fragments carrying additional kinetic energy. It was also found that ρ, the reduced differential cross section: (defined at small scattering angles as $\theta^2\sigma(\theta)$) for peak B and for charge exchange have a maximum at the same reduced scattering angle ($\tau = E\theta$) suggesting that a common primary mechanism may be responsible for both peaks. The experimental results suggest a simple model of the collision which is similar to one used in explaining ion-atom charge exchange in He^+ + Ne and in the non-symmetrical alkali ion-alkali atom cases. In these systems the charge exchange proceeds via couplings between initial and final states that lie close together in energy at large internuclear separation. In He^+ + H_2 a similar situation could exist following the

collisional excitation of peak B at Q = 13.3 eV (as one example). Here as the collision partners separate this state comes close in energy to a number of exchange states resulting in possible transitions to them. After excitation of an exchange state additional interactions with the direct (peak B) and other exchange states occur. In $He^+ + H_2$ the large number of available states results in a dominant role for charge exchange in comparison with the direct scattering. On the basis of this model the peaks of the reduced differential cross sections for the exchange and direct scattering will be found at a common τ value since both processes result from a common primary interaction (the crossing of the incident potential surface with the one resolving to state B at infinite separation).

A similar model accounts for our charge exchange results in the $Ar^+ + N_2$, $Ar^+ + H_2$, and $He^+ + N_2$ cases. Here quasi-resonant processes are possible and couplings between the direct elastic and charge exchange channels occur.

Our studies of the direct and exchange scattering in $Ar^+ + CO$ provide additional justification for the assumed charge exchange model. Fig. 1 shows a typical energy loss spectrum for direct scattering of 2 keV Ar^+ at 0.5^o. Peak A corresponds to quasi-elastic scattering and the two inelastic peaks, B and C, represent electronic excitation of the CO molecule. A fourth peak, 14 volts down from the elastic channel, corresponds to ionization of the CO. At small angles the elastic contributions dominate the scattering. Fig. 2 shows typical energy loss spectra of scattered Ar^o from 1 keV $Ar^+ + CO$ exchange collisions. It can be seen that excitation of the CO^+ ground state ($Ar^+ + CO \rightarrow Ar^o + CO^+(X^2\Sigma^+)$) does occur, but accounts for only a small contribution to the charge exchange. The dominant process

in the scattering involves excitation of the $A^2\Pi$ state of CO^+. The third peak, representing dissociation to $C^+ + O$, is seen to broaden toward higher energy loss with increasing scattering angle, showing that the dissociation products carry off excess kinetic energy. The $B^2\Sigma^+$ state of CO^+ is only weakly excited if at all. The participating direct and exchange states are shown on the energy level diagram of Fig. 3.

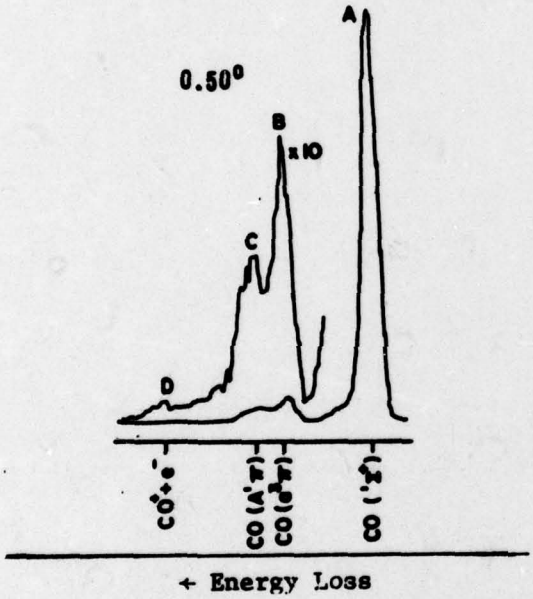

Fig. 1. Energy loss spectrum of Ar^+ from 2.0 keV-0.50° Ar^+ + CO collisions. Calibration of the electrostatic energy analyzer is performed observing known inelastic transitions in Ar^+ + Ar collisions.

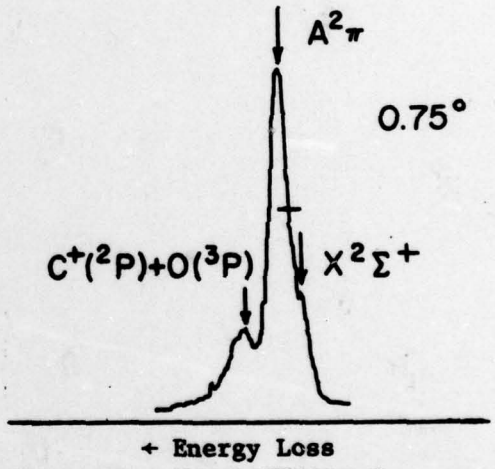

Fig. 2. Typical time-of-flight energy spectrum from 1 keV-0.75° Ar^+ + CO collisions. The state designations are at the positions of the lowest vibrational levels. Quasi-resonant charge exchange would occur at the point of intersection of the short horizontal line with the spectrum.

The experimental results are in good agreement with the model which predicts that the elastic peak A will predominantly populate the $A^2\Pi$ state, and because of a large energy separation the $X^2\Sigma^+$ state will be

weak. The direct inelastic peak B will couple to $Ar^+ + C^+ + O$. The $B^2\Sigma^+$ state in CO^+ is not expected to participate and the spectra show that it does not.

The matching of energy levels in the direct and charge exchange scattering in the ion-molecule collisions studied to date is seen to play an important role in determining the states excited in these collisions. If the direct scattering state is close in energy to a large number of charge exchanged states they should become very important in the collision process. Detailed discussions of the above results have been submitted to A.R.O.D.

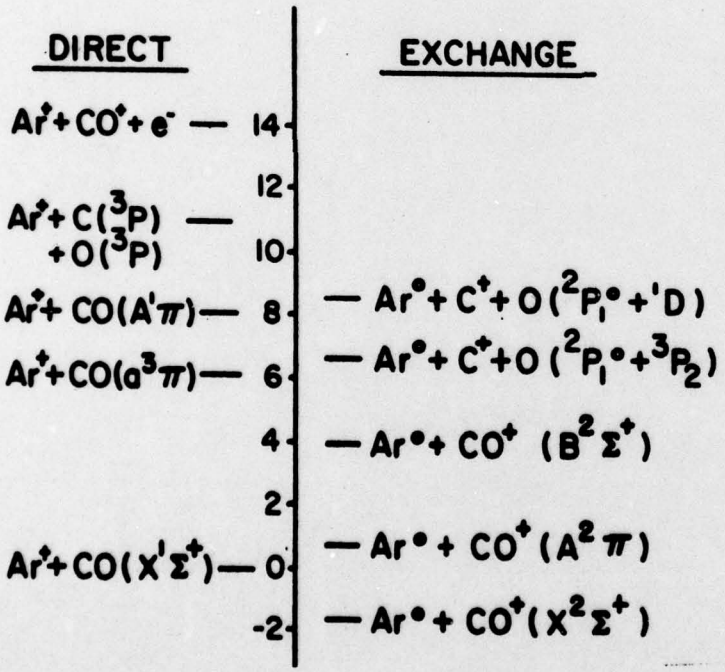

Fig. 3. Selected states in $Ar^+ + CO$. The states seen in direct scattering are identified. The dominant exchange process ($Ar^+ + CO \rightarrow Ar^o + CO^+(A^2\pi)$) results from coupling with the elastic ($Ar^+ + CO\ (X^1\Sigma^+)$ channel. $CO^+X^2\Sigma^+$ is weakly excited because of the larger energy separation from the elastic channel.

B. Scaling Laws

Energy losses were measured in $Ne^+ + D_2$, $Ne^+ + H_2$ and $Ne + D_2$ collisions for beam energies $0.5 \leq E \leq 3.5$ keV and scattering angles $\theta \leq 5$ deg. Rotational and vibrational excitation of the target molecule was found but the probability of direct electronic excitation was seen to be extremely small. The results indicate that the most probable laboratory energy loss T_o for a projectile with mass M_p scattered by a homonuclear binary molecule with atomic masses M scales so that the quantity $f = T_o M/(M_p E\theta^2)$ is a function of the reduced scattering angle $\tau = E\theta$ only, as recently predicted theoretically by Sigmund. The function $f(\tau)$ was found to be the same for the $Ne^+ + D_2$ and $Ne^+ + H_2$ systems, but is strongly dependent on the charge state of the projectile. A complete discussion of these results will be published in the Physical Review and reprints will be sent to A.R.O.D.

C. A detailed study of the direct and exchange scattering in $D^+ + H_2$ is currently in progress.

II. Spectroscopic Studies of Excitation Pathways in Ion-Atom Collisions

A. Polarization Analysis

Total cross sections for the excitation of the $1s3\ell^{3,1}L$ states of atomic helium in $He^+ + He$ collisions were measured in the energy range 60 eV - 1 keV using the technique of photon counting under single-collision conditions. This work was part of the Ph.D. dissertation of David A. Clark ("Optical Polarization Measurements and Excitation Mechanisms in Low Energy Ion-Atom Collisions", University of Connecticut, 1978, unpublished). Electronic excitation in such collisions involves non-adiabatic transitions between electronic states of the diatomic quasi-molecule formed momentarily during the collision. Excitation of a particular "Born-Oppenheimer" electronic level of such a quasimolecule implies that a definite M_L state is excited in the collision. In general, such a state then evolves to a final target or projectile atomic state which is anisotropic (a consequence of the anisotropy of the target atoms with respect to the beam direction). The result is that the optical emission from the separated atomic systems following the collision is often polarized with respect to the ion beam axis. The total cross section in such a case (more common than is generally recognized) cannot be determined by sampling the emitted light at 90^o unless polarization measurements are made along with intensity measurements.

For excitation of target atom states with orbital angular momentum L=0, the polarization is found (theoretically and experimentally) to be zero and the optical emission cross section is proportional to the excitation cross-section for a single (orbital) magnetic sublevel, M_L=0, arising from only a single symmetry, Σ, of the diatomic quasimolecule. For a P-state

excitation with L=1, one can extract the partial cross-sections for the magnetic sublevels M_L=0 and M_L=1 arising from quasimolecular states of symmetries Σ and π respectively. We find, for example in the case of the excitation of the $3\ ^3P \rightarrow 2\ ^3S$ transition in atomic helium with He^+ ions, the partial cross section for excitation of $M_L = \pm 1$ behaves relatively smoothly as a function of energy. On the other hand, the M_L=0 partial cross section shows a clear ("Rosenthal") oscillation pattern as a function of energy, indicating that at least <u>two</u> molecular excited states of He_2^+ of Σ symmetry are forming a <u>closed loop</u> in the excitation pathway, producing quantum oscillations by interference between alternate collision pathways. The separation of the emission cross section data into M_L=0 and $M_L = \pm 1$ partial cross sections separates out the oscillating from the non-oscillating part and identifies the specific substate (M_L=0 in this case) that exhibits interference due to multipath excitation. The Rosenthal quantum-oscillation frequencies involved in the excitation of the 3S, 3P and 3D terms of helium in these collisions are all found to be similar, suggesting that coherent long-range couplings among this whole manifold of states are involved in the excitation process. This situation should be quite common in inelastic heavy particle collisions when several molecular orbitals that are near-degenerate at large internuclear distance R can be coherently excited at small R.

A report on this work has been published (1979) in the IEEE Transactions on Nuclear Science. We hope to extend the present studies to systems in which the valence shell of the target is isoelectronic to helium, e.g. He^+ + Mg or He^+ + Ca collisions, in order to establish the generality of these phenomena.

B. Vacuum Ultraviolet Collision Spectroscopy

We have recently surveyed emission spectra under single-collision conditions in the 500-1100Å wavelength region for several noble-gas, ion-atom collision pairs, in the 8-30 keV energy range. A large number of spectral lines were observed and most were identified. The spectra obtained, for example, from He^+ + Ar and Ne^+ + Ar collisions show that the target state populations are definitely non-statistical. Figure 4 gives a dramatic example of a collisional transfer of an outer-shell electron vacancy in the projectile into an inner subshell (3s) of the target. When the initial vacancy is in a projectile 1s or 2p orbital, transfer to the Ar 3s is enhanced. If the initial vacancy is in the Ar 3p orbital, the process is strongly inhibited. In the He^+ + Ar case, the cross section for excitation of $Ar^+(3s^{-1})$ is large, approximately 10^{-16} cm^2 at its maximum (8.5 keV). Estimates of the charge-exchange cross section from the He^+ projectile to the ground state of Ar^+ are lower than this and a population inversion appears to be possible. Molecular orbital correlation diagrams and energy-level curves as a function of internuclear distance make it possible to estimate which excitations will predominate. Molecular-orbital curve crossing radii are helpful in setting upper limits to the cross sections. These limits are in fact reached in some cases such as the He^+ + Ar case already referred to. In some of the collisions studied, very high states of ionization are observed in the VUV spectra at relatively low collision energy (e.g. an Ar^{5+} line at 588Å is seen in 20 keV Ar^+ + Ar collisions.

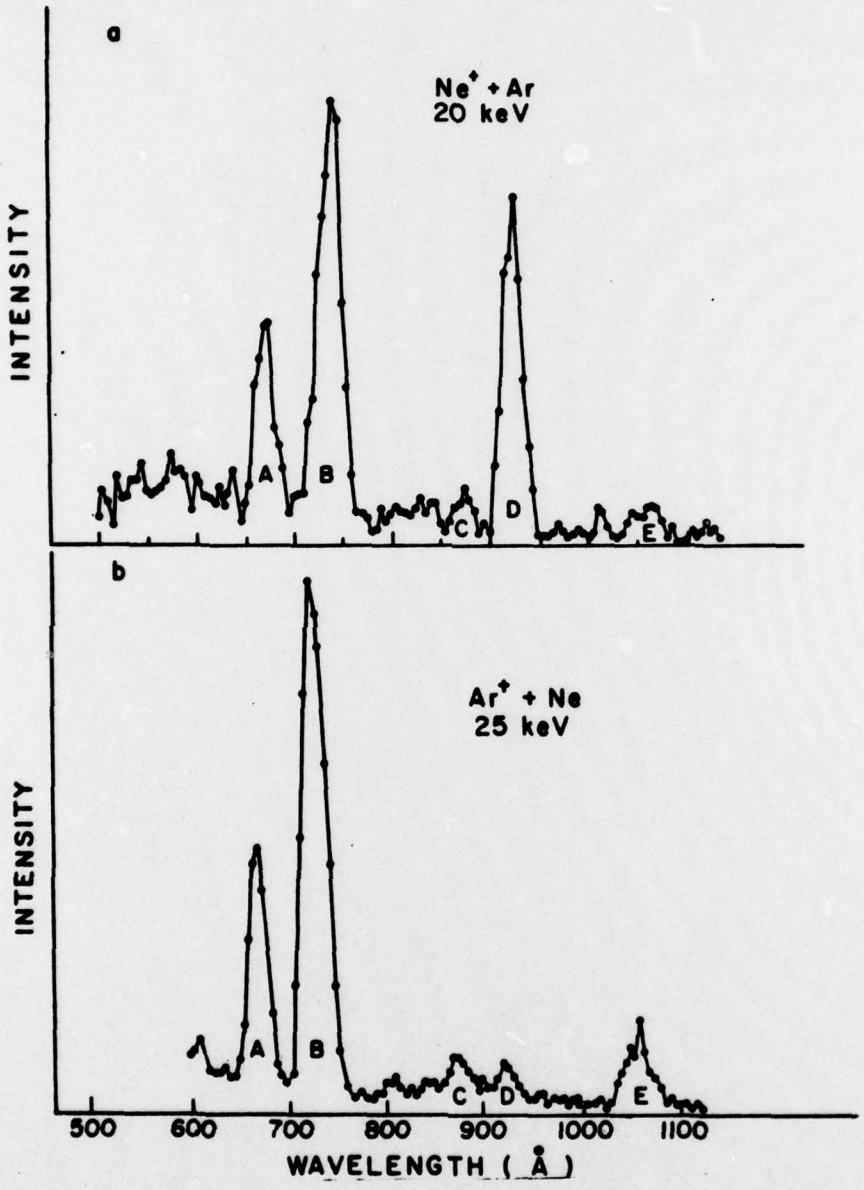

Fig. 4. Comparison of Ne^+ + Ar Vacuum UV Spectrum in the 500-1100Å range with the Spectrum Produced by Ar^+ + Ne Collisions. Note Feature D which consists of the lines at 920 and 932 Å due to 3s vacancies in Ar. Essentially similar spectra were observed with He^+ + Ar (strong excitation of Feature D) vs. Ar^+ + He (very weak excitation of Feature D).

C. Soft X-Ray Spectroscopy from Ion-Beam Collisions with Gases and Solids

A computer-controlled, grazing incidence monochromator has been designed, built to our specifications, and is now in our laboratory. The instrument was designed for high resolution (~ 0.2Å FWHW for Ar-L x rays, λ ~ 56Å) and for relatively high throughput (due to the short focal length, 1m Rowland circle) in order to provide significant new information on ion-induced x rays from single collisions in a gas target. This one instrument covers the 40-3000 Å range with two gratings and weighs less than 200 lbs so that it is readily moved from one ion-beam accelerator to another. Four working accelerators are available in the department to cover the broad energy range from 100 eV - 2 MeV. The spectrometer built for us has now become available commercially.

To date, test spectra have been obtained with the instrument and initial observations of Ar-L x rays from 100 keV Ar^+ + Ar gas-target collisions have been made. We also have some data on 50-100 keV Ar^+ collisions with graphite surfaces that suggest some possible new methods for the characterization of solid surfaces, especially contaminated surfaces and those that have been altered by ion implantation. Following 100 keV Ar^+ implantation of the graphite, strong Ar_L x-ray production is observed when the ion beam irradiation is continued. After prolonged irradiation and sputtering of the surface by the Ar^+ beam, the C_K line appears. A possible explanation (L. Feldman, private communication) involves the doping of the first 1000-2000 Å of the surface with implanted argon. Incident Ar^+ projectiles then acquire 2p vacancies from Ar^+ + Ar collisions. The $Ar^+(2p^{-1})$ then transfers the vacancy via rotational coupling to the carbon host 1s shell giving a C_K x ray. If this model

proves to be correct, the spectrum becomes sensitive to the ionic state of the projectile within the solid.

A different experiment involved measurements of the total Ar^+ ion-induced yield of soft x rays at 100 keV for the graphite target which had previously been implanted at low energy (50 keV) with P^+ ions. This near-surface low energy implant resulted in an x ray yield that was larger by approximately an order of magnitude than the yield from a deeper implant at 100 keV. These observations are consistent with the possibility that we may be seeing soft x ray "SCANNIR", i.e., that a part of the x-ray yield comes from implanted ions that have been sputtered off the surface in highly excited states. A careful study of the spectrum at the high resolution possible with grazing incidence should confirm whether the x rays are from isolated ions or from within the solid. We appear to be seeing effects in the soft x-ray spectrum that are quite sensitive to both the contamination to the surface and to the depth distribution of implanted ions. Similar studies of ion beam interactions with a variety of metallic surfaces are planned.

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Manuscript in Preparation

Hodge, W.L., Jr., F.W. Martin, E. Pollack and W.W. Smith. K x-ray spectra in carbon compounds following collisions with MeV oxygen ions. (to be submitted to Physical Review).

Degrees Awarded to Recipients of ARO Support

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