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FAST ION IMPACT ON THIN METAL FOILS:
IMPLICATIONS OF THESE DATA FOR DEVELOPMENT
OF TRACK STRUCTURE MODELS

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Electron Emission Resulting from Fast Ion Impact on Thin Metal Foils: Implications of These Data for Development of Track Structure Models

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Abstract:

Experimental information useful in improving and testing theoretical models of energy deposition and redistribution in particle-condensed phase media is discussed. An overview of information available from previous, and ongoing, studies of the electron emission from thin foils is presented and the need for doubly differential electron energy and angular distribution data stressed. Existing data are used to demonstrate that precisely known, and controllable, surface and bulk conditions of the condensed-phase media are required for experimental studies of this type. Work in progress and improvements planned for future studies at the Pacific Northwest Laboratory are outlined.

Introduction:

Energetic particles traversing matter undergo coulomb interactions with bound target electrons. These interactions deposit energy in the target media at localized sites having nanometer dimensions. Energetic electrons produced in ionizing collisions at a particular site can redistribute a portion of the deposited energy over a larger area via subsequent interactions

with other bound media electrons. In each interaction, media atoms or molecules are excited and will therefore react with their neighbors in a different manner than they do when in their relaxed states. This can produce "chemical" changes within the media.

Modeling the effects evolving from energy deposition is therefore rather complex. It involves knowledge about the products generated in individual projectile-media interactions, about emitted electron-media scattering and ionization processes, and about how excited/ionized target atoms and molecules relax or interact with their neighbors. As both the impinging projectile and the emitted electrons lose energy and slow due to interactions with the media, information about these processes is required for a wide range of energies. An added complication is that in finite sized media some of the ionized electrons may reach the surface and escape into the vacuum. This removes some of the originally deposited energy from the media.

Understanding the energy deposition and redistribution processes is vitally important in the fields of radiation induced damage to biological tissue and materials. Information about electron transport and emission is essential for designing systems where plasma-surface interactions occur, e.g., in thermonuclear reactors or in gas discharges, or where surface charging effects play a role. Electron emission from surfaces also plays a significant role in areas where accurate measurements of beam currents are required or for situations

where particle detectors must be designed or used.

Modeling needs:

The philosophy behind our studies at the Pacific Northwest Laboratory is that models used to describe energy deposition processes require two types of experimental information. First, and foremost, they require as detailed information as possible about individual particle-media interactions. These data constitute the input parameters upon which the model is constructed. Secondly, and of equal importance, they require accurate experimental measurements for testing assumptions required when "other than ideal" systems are modeled, i.e., data are required to ascertain the reliability of the assumptions and methodology used.

The type of information required as input parameters is illustrated in Fig. 1 where an impinging particle, P , possessing charge, z , and velocity, v_p , interacts with a target, T . Generally a single electron is ejected; its velocity and direction with respect to the incoming projectile velocity being v_e and θ respectively. A target ion, capable of "chemically" interacting with its neighbors or recombining with other electrons or ions, is also produced. After the collision the ion charge is z' , which may be the same, larger, or smaller, than z . The final ion velocity, v_p' , is always smaller than v_p . This constitutes the original energy deposition process.

A partial redistribution of the deposited energy occurs when

the ejected electron undergoes subsequent elastic and inelastic interactions with other bound electrons in the media. Elastic collisions only alter the direction of travel whereas inelastic collisions produce additional ionization of the media. These inelastic processes are identical to those diagrammed in Fig. 1 with, of course, the modification that $z' = z = -1$. Such processes continue until the original particle and all the ejected electrons come to rest or escape the media.

It should be mentioned that although energy deposition processes typically are expressed by a single electron being ejected to the continuum, i.e., single ionization, gas phase studies¹ have shown that the ejection of additional electrons is sometimes quite probable. Gas phase studies have also shown² that electron emission from interactions involving an increase or decrease of the projectile charge can also be quite important. For modeling purposes, multiple ionization or z changing interactions may be particularly relevant since they deposit different amounts of energy at localized sites and most likely produce different target ion species. This emphasizes that energy deposition models should treat the process in a stochastic rather than in an "average" manner.

Thus modeling the energy deposition may be cumbersome, but it is a very straightforward procedure once one has sufficient information about the individual particle-media interactions. Unfortunately, within a condensed-phase media these individual interactions simply cannot be experimentally investigated in such

detail. Therefore a common method has been to use information obtained from gas-phase energy deposition measurements and to add appropriate modifications to account for bulk properties that are absent in the gas-phase data, e.g., to use condensed-phase differential oscillator strength information to modify the gas-phase cross sections in the low-energy regime. (see, for example, ref. 3). These modified data can then be used as input parameters and the modeling process proceeds as usual. But this requires various assumptions and the validity of those assumptions needs to be tested.

Testing the assumptions requires information about electron production and transport in condensed-phase media, which implies that measurements of the electron emission from the media can be used for this purpose. However the complication is that electrons are produced at varying depths within the solid and hence they suffer varying degrees of energy degradation as they travel toward the surface. Also they must be transmitted through the solid-vacuum interface in order to be observed. Hence the measured electron intensities are a convolution of the electron production, electron transport and the electron transmission probabilities associated with the media and all these pieces of information may not be available or easily obtainable for the particular media of interest.

A method of circumventing this problem is to test the model and its assumptions by studying "ideal" condensed-phase systems, meaning systems that can be modeled with sufficient accuracy and

also be experimentally interrogated under well specified conditions. Sufficiently detailed measurements of the electron emission from such systems then provides a means of testing energy deposition models and may also provide insights into improving the models. To date we are aware of no detailed comparisons of this type, primarily because accurate experimental data are lacking.

Experimental work designed to supply these needed data is in progress at the Pacific Northwest Laboratory. However, prior to describing our studies, we will first provide a general overview of what is already known from previous electron emission studies. Due to limited space and available data, we will concentrate on electron emission resulting from fast ion (typically proton) impact on thin metal foils. We wish to emphasize that the examples cited are chosen primarily because we are more familiar with these studies; we do not intend to imply that they represent the only investigations in this field.

Previous Studies:

Experimental studies of electron emission from condensed-phase targets are typically performed using thin, self-supporting metal foils in order to ensure that the impinging proton energy remains roughly constant during transit. As will be shown, the electron emission is extremely sensitive to the surface conditions, so only data involving reasonably clean or known surface conditions will be discussed here.

The most prevalent studies are of the total electron yield per incident particle, γ ; ⁴⁻⁸ some studies of the forward versus backward emission, γ_f and γ_b have also been performed. ⁹⁻¹² More detailed information is available from electron energy, ^{5,10,13-14} $N(\epsilon)$, and angular, ¹⁵⁻¹⁶ $N(\theta)$, distribution measurements. However, for testing purposes we strongly believe that doubly differential energy and angular distributions, $N(\epsilon, \theta)$, are required since they will provide the most stringent tests. For example, total electron emission studies may provide certain insights into the overall process but using them for testing purposes is similar to evaluating various mathematical descriptions of a square and a triangle by only comparing their calculated surface areas with that of a test object. More highly differential tests are obviously required.

A few studies of $N(\epsilon, \theta)$ have been reported for "clean" surface conditions ^{8,16-23} but these rarely include full angular and energy information for the emitted electrons. From these and other studies, information about how the electron emission depends on the proton velocity, v_p , target z , target temperature and surface conditions is available.

a) Gaseous vs. condensed-phase electron emission:

Since modified gas-phase data generally are sometimes used to provide the ionization probabilities in track structure codes, we begin by comparing the electron emission from gaseous and condensed phase targets. The only direct comparison of gas- and

condensed-phase electron emission data of which we are aware is shown in Fig. 2. It is well known that bulk properties influence the production of low energy electrons and that impurities on the foil surfaces strongly influence the transmission of low energy electrons but these influences are believed to play minimal roles for higher energy electron emission. For that reason, Toburen²⁴ felt justified in comparing the electron emission from a thin carbon foil obtained using standard high vacuum conditions (approx. 10^{-6} Torr) with the emission obtained from a tenuous methane gas target.

Because of uncertainties in the foil thickness, the carbon foil cross sections are uncertain within a factor of two. However the important differences between the gas- and condensed-phase data are a suppression of low energy electrons and a dramatic increase in the high energy electron production observed for the foil target. These are attributed to absorption of low energy electrons and scattering of forward directed high energy electrons to larger angles, respectively. Contamination of the foil surfaces and bulk effects undoubtedly also influence the electron emission.

b) Electron emission from thin foils:

We now discuss general features and what is already known about electron emission from thin foils. From an experimental viewpoint, the most important point in such studies is to provide a clean surface since the electron emission is extremely

sensitive to surface contaminants residing on the surface. Surface contamination can be a combination of oxides chemically attached to the surface or adsorbed and occluded gases from the residual vacuum in the experimental apparatus. These contaminants tend to lower the work function of the surface;²⁶ thus as they are removed the emission of lower energy electrons decreases. (see Fig. 3) As can also be seen, removal of these impurities causes the maximum in the electron yield to shift to higher energies and the total yield of electrons to decrease dramatically. In addition, the width of the low energy maximum increases with the removal of impurities.¹³ Therefore, unless the surface conditions are extremely well specified and controlled, comparisons between experimental data and theoretical predictions are meaningless.

With this in mind, the following generalities are based upon studies performed where the surfaces were quoted as being "clean". In many cases this was defined as "less than 0.3 monolayer".⁸ Beginning with the total electron emission, for proton impact it has been shown,^{4,7} as expected,²⁷ that the total yield of electrons is proportional to the stopping power of the primary ionizing particle. For modeling purposes, however, it is important to remember that γ is target and projectile dependent⁹ and that γ increases in magnitude dramatically as the surface becomes contaminated.

Investigations of the forward and backward electron emission yields for fast protons impacting on thin carbon foils have

shown¹¹ that approximately 15% more electrons are emitted in the forward (direction of exiting proton) than in the backward direction. Since low energy electron emission is the primary contributor to γ , these features may prove to be useful in testing assumptions about condensed-phase modifications to existing codes.

These forward/backward emission studies might also be used in a more unique fashion. For example, information about the range and ionization induced by electrons traversing condensed-phase media has been obtained from investigations of the forward and backward electron emission yields using fast ionic projectiles containing loosely bound electrons, e.g., H^+ .¹¹ Since the loosely bound electron is stripped from the projectile within a short distance after entering the foil, it can scatter back in the direction of the incoming beam or can travel in the forward direction causing additional ionization. If the foil thickness is less than the range of this electron, it, as well as any additional electrons produced along the way, will exit the foil. This increases γ_f as shown in Fig. 4. For thicker foils, these electrons cannot escape and the yields for H^+ and H^+ impact are identical. Since the stripped electron initially has the same velocity as the incoming beam, varying the beam energy offers a unique method of studying electron transport in foils.

Investigations of the low energy portion of the emitted electron energy spectra have shown that the shape is a function of the target material.¹³ However for light ion impact, the

position where the emitted electron yield maximizes and its width were found to be independent of the projectile impact velocity. For heavier projectile impact, the situation is different. The emitted electron energy distributions were wider and shifted to higher electron energies. Also the widths of the distributions increased with impact velocity for heavy ion impact. These changes as a function of projectile mass or velocity almost certainly are the result of differences in the initial energy deposition processes; hence they might be useful in testing this aspect of models.

The angular dependence of the emitted electrons might also be useful in testing the initial energy deposition processes since a $\cos\theta$ dependence (relative to the surface of the foil) is expected for isotropic source conditions. This dependence has been confirmed¹⁷ for higher energy electron emission but, as will be shown later, it does not apply for low energy electron emission.

For testing theoretical models, we believe that doubly differential electron emission measurements should be used. However, from an experimental viewpoint, it is essential that these data be obtained under well specified conditions. The carbon foil data shown in Fig. 2 were obtained under standard high vacuum conditions where surface contamination was present but it was assumed that the higher electron energy data would be unaffected by any contamination. In Fig. 3, low energy electrons were indeed shown to be strongly influenced by surface

contamination.

Fig. 5 demonstrates this again for zero degree electron emission from sputter cleaned and untreated copper foils.²³ Fig. 5 also demonstrates that removal of surface contaminants also leads to an increase in the yield for rather energetic electrons, electrons having velocities as large as the projectile velocity in the example used. This increase cannot be explained in a simple manner but may have ramifications concerning theoretical models.

The zero degree electron emission data in Fig. 5 also contain a strong convoy electron peak at approximately 650 eV. These convoy electrons might prove to be useful in providing information about electron transport and energy degradation in condensed-phase media since they are born within the media with rather well defined velocities and then suffer inelastic interactions prior to escaping the media.¹⁹ Additional information about convoy electrons is contained in references 18-21.

Fig. 5 also demonstrates that the differential electron emission depends on the target temperature. A close inspection of the data indicates the low energy electron emission decreases with decreasing target temperature, a feature that again emphasizes that experimental conditions must be extremely well defined if the data are to be useful for testing purposes.

Present work:

These studies have demonstrated that the electron emission is strongly influenced by surface and bulk properties and that low energy electron emission is particularly sensitive to bulk properties. Thus experimental conditions, particularly regarding low energy electron emission, must be extremely well known and controlled in order to provide data capable of testing and improving energy deposition models. The lack of doubly differential electron emission data of sufficient quality for testing track structure models is currently being addressed at PNL. We are studying the energy and angular distributions of electrons emitted from thin foils subjected to fast proton irradiation.

Initially emission from thin carbon foils is being studied since self-supporting carbon foils are readily available commercially and other experimental information for proton-carbon foil systems exists (see above). Electron time-of-flight (TOF) techniques are employed since they are particularly well adapted to studying low energy electron emission. Likewise extreme care is taken to avoid stray electric and magnetic fields within the target and spectrometer regions which may influence the low energy electron emission.

Unfortunately, our initial studies were hampered by surface cleanliness conditions. We had anticipated that the foils could be sputter cleaned in a similar manner to that used previously.⁸ But we were unable to achieve reproducible emission spectra which

means that either our technique was in error or the electron emission at a specific angle is more sensitive to surface contamination than is the total yield of electrons. Our initial studies also suffered from restricted base (and operating) pressures of $1-2 \times 10^{-9}$ Torr arising from a damaged sealing surface. These limitations are currently being addressed in the next phase of the work.

Data presented here were accumulated in the following manner. Energy and angular information were obtained by interacting a pulsed proton beam (intensity $< 10^{-11}$ A) with a thin ($5\mu\text{gm}/\text{cm}^2$) carbon foil and measuring the emitted electron flight times, i.e., energies, using a rotatable detector. Experimental parameters allowed us to measure electron energies up to 100 eV with reasonable accuracy and higher energies with ever decreasing accuracy. Emission angles relative to the proton beam that could be investigated were $15-75^\circ$ and $105-155^\circ$, the foil surface being normal to the proton beam. Additional details are available in ref. 28.

Since our attempts to sputter clean the foils using the unpulsed proton beam were unsuccessful, we heated the foils to between 200 and 300 degrees C for approximately 24 hours in order to remove adsorbed gases. However, we had no direct method of confirming the surface cleanliness that resulted. Rutherford backscattered protons indicated a 2-3% oxygen contamination of the foil after baking; whether this represents surface or bulk contamination is uncertain.

After baking, the foil was removed from the oven and a TOF spectrum accumulation began within a few seconds. Sufficient statistics were achieved within approximately 100 seconds, the data were stored, and a new spectrum was measured. Using this procedure, we obtained "snapshots" of how the buildup of surface contamination influences the electron emission. Results are shown in Fig. 6.

Two major features are seen. One is that surface contamination causes the low energy electron emission to increase in magnitude which broadens and shifts the energy distribution to lower energies. The shift is in agreement with the electron emission data integrated over all angles as shown in Fig. 3 but the broadening is opposite to that shown. The second feature is the decrease in the yield for high energy electrons with increasing surface contamination. This is in agreement with data shown in Fig. 5.

Assuming unit sticking coefficients, the buildup of O_2 contamination on the surface was calculated and is shown in Fig. 6. The assumption used is that the surface was initially clean, although this could not be directly verified. Nevertheless, the data clearly indicate that doubly differential emission is sensitive to even a few percent of a monolayer of surface contamination. If subsequent experiments using foils where we are absolutely certain of the initial surface conditions confirm this, it casts doubt upon the surface conditions used in previous measurements. For example, Hasselkamp et al.⁴ stated that they

sputter cleaned their targets and interrogated them within 30 minutes. With their quoted base pressure of 5×10^{-9} mbar, this implies a surface contamination of roughly half a monolayer. Burkhard et al.⁸ claim their surfaces to be clean to better than 0.3 monolayers of contamination.

Fig. 6 demonstrates that such levels of contamination seriously alter the low energy electron emission yields which should also effect γ . We stress, however, that our findings may be subject to our initial surface conditions which we are unable to specify. We also point out that we may be observing a temperature dependence similar to that shown in Fig. 5 since the foil was cooling from oven to ambient temperatures during spectra accumulation. We doubt that temperature plays a role, however, since temperature dependent features extend⁸ to higher electron energies than where we observe changes in the spectra.

In Fig. 7 we show the electron emission yields as a function of angle of emission. The foil was again baked as described above, but a comparison of the 135° data in Figs. 6 and 7 shows differences in the time zero spectra. If the initial surface conditions in Fig. 6 are indeed contamination free, the comparison implies that approximately 0.17 monolayer of (presumably) oxygen contamination is initially present in Fig. 7. Assuming this to be the case, the buildup of contamination from background gases is again calculated and the total contamination is tabulated in Fig. 7.

Also shown are the relative intensities as expected from a

$\cos\theta$ angular dependence. For electron energies above 30 eV this $\cos\theta$ dependence appears to be correct; however lower energy electron emission exhibits a much stronger dependence. This may arise in part from the suppression of low energy electrons emerging more parallel to the foil surface.

Thus, the present data provide two new pieces of information. They indicate that surface contamination levels above a few percent of a monolayer strongly influence the low energy electron emission yields and that the electron emission stabilizes when the surface contamination is between approximately $1/3$ and 1 monolayer. Secondly, they show that a $\cos\theta$ angular dependence does not apply for low energy electron emission.

Summary and suggestions for future studies:

Experimental data required for modeling energy deposition and transport in condensed-phase media have been discussed. For testing purposes, measurements of the electrons emitted when thin metal foils are bombarded by energetic particles are desirable. Past studies were discussed in order to show what is already known from such studies and the need for detailed electron energy and angular distribution information is stressed. Experimental work in our laboratory designed to provide such information was discussed.

The data presented emphasize that electron emission measurements useful for testing energy deposition models must be

obtained under extremely well controlled, and defined, surface and bulk conditions. Hence, additional studies are in progress in our laboratory where improved vacuum and surface cleaning procedures will provide known, and reproducible, surface conditions. A combination of electron time-of-flight and electrostatic energy analysis will be used in order to investigate the entire emitted electron spectrum. Simultaneous multiple-angle detection of the electrons is planned in order to investigate the angular and energy distributions under identical target conditions.

Future studies will employ condensation of gases on a cooled surface or *in situ* vaporization of metals on a clean surface to investigate electron production and transport in precisely known condensed-phase media. These improvements are intended to provide data having sufficient quality and details that should provide the first real test of track structure models.

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Figure Captions:

- Fig. 1. A schematic diagram of an individual ionizing interaction where initially the projectile, P , has energy, T , and charge, q . As a result of the interaction, the projectile energy decreases slightly to $T - \delta$, and its charge changes to q' , where q' may be larger, smaller, or equal to q . An electron is ejected with energy ϵ and direction θ and an excited target, T^* , is produced.
- Fig. 2. Doubly differential electron emission cross sections compared for 1 MeV protons interacting with a tenuous methane target and a thin carbon foil, and — , respectively. The data are taken from ref. 24.
- Fig. 3. Electron yield, $N(\epsilon)/\text{incident ion}$, as a function of emitted electron energy and surface cleanliness for 500 keV $\text{Ar}^+ - \text{Be}(0)$ interactions. Also listed are the total electron yields, γ . Data are taken from ref. 13.
- Fig. 4. The forward electron emission, γ_f , for H atoms relative to that for protons passing through carbon foils. The curve serves only to guide the eye. Data are from ref. 11.

Fig. 5. Zero degree electron emission spectra for protons impacting on sputter cleaned and untreated copper foils. The left and right sides of the figure are obtained for different target temperatures. v_B is the Bohr velocity. Data are from ref. 23.

Fig. 6. Electron yields measured at 135° with respect to the beam direction for 2 MeV protons interacting with a thin carbon foil. The spectra were taken as a function of time after the foil was removed from an oven used to remove surface impurities. Assuming initially clean surface conditions, the buildup of O_2 on the surface was estimated.

Fig. 7. Electron yields as a function of angle of emission for 2 MeV proton-carbon foil interactions. The spectra were obtained at different times after removal from an oven used for cleaning the surfaces. Comparing the initial spectra with that in the previous figure indicates the initial level of surface contamination is approximately 0.17 monolayer. Additional O_2 buildup from the residual vacuum is calculated and shown. Also shown are the relative intensities expected from a $\cos\theta'$ dependence where θ' is measured normal to the foil surface.

Ionizing Interactions

Before

$P(T, q)$

After

$P(T - \delta, q')$

$e(\epsilon, \theta)$

Fig 4

Fig 5

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