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Abstract

Energetic heavy ion beams are capable of enhancing the adhesion of metallic films to a variety of substrates. Gold films (200-600 Å) evaporated onto substrates of tantalum and silicon (with native oxides) were bombarded with ions of ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{58}Ni at 2.85 MeV/nucleon. The threshold dose required to produce a peel strength greater than the Scotch-tape peel strength for a gold surface was measured as a function of ion species and angle of incidence. We observed the threshold dose to vary as the cosine of the angle with respect to normal incidence. The dependence on particle type for the Au-Ta system (with approximately 40 Å native oxide) was found to be $D^{th}(cm^{-2}) = 10^{17} \left(\frac{dE}{dx}\right)_{Au}^{-3.0\pm.02}$ for all particle beams where $\left(\frac{dE}{dx}\right)_{Au}$ is the electronic stopping power (MeV cm^2/mg) of the ion in gold. A substrate of 6000 Å of tantalum oxide gave identical results. The value of the exponent, -3.0 ± 0.2, differs significantly from the value -1.6 ± .02 reported earlier by Tombrello, et al., for Au-Ta. The Au-Si system is described by $D^{th} = 6 \times 10^{18} \left(\frac{dE}{dx}\right)_{Au}^{-4.1\pm.03}$. Auger analysis of the Au-Ta interface suggests a migration of the native oxygen as a result of ion bombardment. For the Au-Ta system, some irradiated regions that passed the Scotch-tape test just after bombardment were observed to fail when the test was repeated a few days later. Thicker gold films and higher doses resulted in longer adhesion times. These and other observations suggest that the post-irradiation decline of adhesion for Au-Ta is caused by diffusion of ambient atoms through the gold film rather than by an intrinsic time dependence of the adhesive forces.

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I. INTRODUCTION

The observation that energetic ions passing through the interface of a thin metallic film and an insulating substrate could significantly improve the adhesion of the thin film[1)] has led to a variety of experimental investigations of this general phenomenon.[2-8)] In addition to the possible practical applications, interest in this subject grew because of the idea that the mechanism for enhancing adhesion was different than the ion beam mixing mechanism associated with ions of low energy. In the latter case, the ions cause a physical displacement or mixing of atoms at the interface through direct nucleus-nucleus collisions, i.e. through the nuclear stopping mechanism. However, ions whose energies are sufficiently high that the main interaction is with the electrons in the stopping medium would impart little momentum to the atoms. Any atomic displacement would only occur subsequent to the ion's passage and through secondary processes. Examples of the latter are the ion-explosion[9)] or thermal spike mechanisms,[10)] which lead to track formation and sputtering in insulators. These mechanisms depend on the long electronic relaxation times in an insulator, and thus are not expected to be present in a conducting or semiconducting medium. It is possible, however, that the observed bonding of films to metal substrates is due to the native oxide layer that exists at the interface of nearly all the systems studied to date.

The basic questions concerning the mechanisms of adhesion enhancement remain open even though a number of well-conceived experiments have provided important information. These questions concern:

(i) the relative roles of atomic displacement, electronic bonding and physical adsorption in producing enhanced adhesion.

(ii) the thickness of the interface region.

(iii) the importance of an oxide layer in the case of conducting or semi-conducting substrates.

(iv) the dependence of adhesion on post-irradiation conditions (annealing, ambient gases, for example), and

(v) the permanence of the adhesion enhancement.

The present experiments began with the object of studying the size of the interface through variation of the angle of the bombarding ion with respect to the sample. These measurements then led us to examine the dependence of adhesion enhancement on the type of projectile for both the Au-Ta and Au-Si systems. Given the importance of surface preparation and condition in determining adhesion, an analysis of the Au-Ta interface at irradiated and unirradiated areas was made with a scanning Auger microprobe. This analysis revealed the typical 30-40 Å layer of native oxide present at the surface of the tantalum. (Although for convenience we shall refer to the samples as Au-Ta, it will be shown that the interface at which adhesion takes place is between the thin film and the oxide layer.) The Auger analysis also indicated, as a result of bombardment, a migration of the oxygen at the interface into the substrate. The permanence of the adhesion enhancement was studied by repeating peel tests at regular time intervals after irradiation.

The following sections present the above mentioned experiments and results. A discussion of the results is given in the last section.

II. Au-Ta SAMPLE PREPARATION AND IRRADIATION CHAMBER

Tantalum substrates were cut from cold-formed sheets 0.5 or 1.0 mm in thickness.[11] They were cleaned in hot detergent (Alconox), rinsed in de-ionized water, etched in a 1:1 solution of nitric acid and water, and rinsed again in water. A rinse in reagent-grade ethanol preceded loading the samples into the evaporator. Following a glow-discharge cleaning in commercial grade argon, between 200 and 600 Å of gold were deposited on the substrate at a pressure in the range of 2×10^{-6} Torr to 8×10^{-6} Torr. The gold films could be removed by applying Scotch-tape (no. 810) to the surface and peeling it with a force of about 5000 dynes per cen-

timeter of tape width. Most of the measurements were made with samples of this type. Samples that had been sputter cleaned prior to evaporation (by lowering the argon pressure from 200 microns of Hg to 90 microns and by raising the discharge voltage from 650 to 1600 volts) had sufficient initial adhesion to pass the Scotch-tape test (i.e., peeling the tape from the sample left the gold film attached to the substrate). The gold film could be removed, however, by gently abrading the surface with a cotton bud. Certain areas of the Au film, e.g., near the edges of the sputter cleaned samples, could not be removed even with a vigorous rubbing with a laboratory tissue or a finger.

The samples were irradiated in the chamber shown schematically in Fig. 1. The samarium-cobalt magnets suppressed secondary electrons while the Faraday cup surrounding and electrically connected to the sample ensured complete charge collection and integration. The sum of the current on the collimator and on the sample equalled the current measured in another Faraday cup located at a waist in the beam about 12 meters upstream from the target. In order to obtain uniform irradiation a ramped steering magnet (beam wobbler) was used to sweep the beam across the 2mm diameter hole in the collimator.

III. THE DEPENDENCE OF THE THRESHOLD DOSE FOR ENHANCED ADHESION ON THE ANGLE OF INCIDENCE

The angle of incidence was varied by rotating the sample while leaving the collimator aligned normal to the beam. For a given number of particles through the collimator, the dose (particles per square centimeter of interface) was thus proportional to cos (θ), where θ is the angle of the beam with respect to normal incidence. Irradiations were made for a given angle at a series of locations on the sample by varying the number of particles through the collimator in steps of $\sqrt{2}$. Next to each of these locations was an area irradiated at normal incidence by the same number of particles through the collimator. All irradiated areas were tested by a single strip of tape removed in a single peel, so that the dose required to achieve adhesion at any angle could be measured quite accurately relative to the threshold dose at normal incidence.

The irradiations were made with a beam of 2.85 MeV/nucleon ^{35}Cl ions provided by the Pelletron accelerator at the Weizmann Institute. Figure 2 shows an example of a peel test on a Au-Ta sample irradiated at 0° and 80°.

The result of the measurements at different angles was that the number of particles through the collimator that produced an enhancement just sufficient to pass the Scotch-tape test was independent of the angle of incidence. Since the area illuminated varied as $(\cos\theta)^{-1}$, the threshold dose varied as $\cos\theta$. The results are shown in Fig. 3. A measurement comparing the threshold dose at normal incidence and at 85° was also made using the sputter cleaned sample with high initial adhesion. In this case the abrasion test using a cotton bud was applied, and the results were the same as obtained with the peel test.

A comparison of the adhesion enhancement for a gold film on a silicon substrate was made at normal incidence and at 80° with a Ni beam. Again, the threshold dose varied as cos (θ). This suggests that the form of the angle dependence is a general feature, independent of the film-substrate combination and the particular test for adhesion.

Since, for a given integrated charge, the size of the irradiated area is larger and hence more convenient to work with, subsequent measurements of the threshold dose for different beams and film-substrate combinations were made at angles varying from 60° to 85°. Using an angle of 85° reduces the number of particles required to enhance adhesion over a given area by over an order of magnitude (a point of practical significance). The ranges of the ions used in these studies are sufficiently long that, even at 85°, the ions come to rest at a point in the substrate far away (about 1 micron) from the interface.

The threshold doses quoted in this paper represent the equivalent dose for normal incidence, unless specifically noted otherwise.

IV. THE DEPENDENCE OF THE THRESHOLD DOSE ON THE BOMBARDING ION

The threshold doses for the 200 Å Au-Ta samples used in the above measurements were typically 1-2 $\times 10^{14}$ cm^{-2} for 99 MeV ^{35}Cl (dE/dx = 6.4 MeV/cm^2/mg in Au, ref. 12). This is 5 to 10 times the dose anticipated from the results for the 500 Å Au-Ta samples bombarded at Caltech[4)] and 2.5 to 5 times larger than found for similar 500 Å samples irradiated at Lawrence Berkeley Laboratory.[2)] While it is generally known that the level of adhesion can depend very sensitively on the surface preparation, we have no specific explanation for this difference. The cleaning procedures were similar to those described by Mendenhall[2)]. (One difference, however, was our use of a glow-discharge cleaning prior to evaporation; this is discussed in the last section.) Therefore it was decided to use other ions in addition to ^{35}Cl to determine whether the dependence of the threshold dose in our case would still vary as $(dE/dx)^{-1.6}$ even though the absolute dose was higher. The same power, -1.6, had been observed in the irradiations made at Caltech[4)] and at Berkeley,[2)] even though the absolute doses differed systematically by a factor of two.

A. Tantalum substrates

Beams of ^{12}C, ^{16}O, ^{28}Si and ^{58}Ni at energies of 2.85 MeV/nucleon were used. The measurements were done on a total of three different samples of 600 Å Au-Ta whose threshold doses for ^{35}Cl were 2.4, 3.3, and 3.4 $\times 10^{14}$ cm^{-2} respectively. (The higher doses here arise from the thicker gold film, see section VII.B). The use of a Cl beam on each sample in addition to one or two other particle types permitted us to normalize the threshold dose for any beam to that for Cl. The results are listed in Table I and are shown in Fig. 4. They are well described by the relation

$$D^{th} = 10^{17}(dE/dx)^{-3.0\pm0.2}\ cm^{-2}$$

where dE/dx is the stopping power in gold in MeV/cm^2/mg. As found in previous results[2,4)] also shown in Fig. 4, the threshold dose follows a power law in dE/dx. Quantitatively, how-

ever, our result for the exponent, -3.0 ± 0.2, is very different than that obtained earlier, -1.6 ± 0.2. We are confident that this difference does not arise from the method of charge integration in either experiment nor from the application of the Scotch-tape test. Furthermore, it is unlikely that our use of a somewhat higher bombarding energy is responsible. (The energies used in ref. 2 varied from 0.1 to 2.2 MeV/nucleon).

To check whether 40 Å of native oxide was already thick enough to render the metallic Ta substrate irrelevant, we prepared a sample with a very thick oxide layer (about 6000 Å) by heating it at 450° C in air for 10 hours. We then subjected it to exactly the same cleaning procedures before evaporation. The threshold doses observed for Ni and Cl beams were the same within experimental error as those found for the regular Au-Ta samples.

B. Silicon substrates

The silicon substrates were 1000 ohm-cm n-type wafers with a highly polished surface, which should have a native oxide layer of the order of 20-30 Å. The samples were cleaned in hot detergent and then rinsed in ethanol before being placed in the evaporator. A glow-discharge cleaning preceded the deposition of 250 Å of gold. The results obtained with Si, Cl and Ni beams are given in Table II and in Fig. 4. In the range of stopping power covered by these three beams, the threshold dose is described by

$$D^{th} = 6 \times 10^{18} (dE/dx)^{-4.1 \pm 0.3} \text{ cm}^{-2}$$

where dE/dx is for ions stopping in gold.

C. Silicon dioxide substrates

The silicon dioxide substrates were of fused silica, cleaned in hot detergent, and etched in a solution of KOH for one minute before the rinse in ethanol. Following a glow-discharge cleaning, 240 Å of gold was evaporated. It was possible to enhance adhesion with beams of Si, Cl and Ni. However, the determination of the threshold was not as clear and unambiguous as with the tantalum and silicon substrates. In the latter two cases there was always a sharp

demarcation where the gold film either tuck to the substrate over a continuous portion of the irradiated area or was completely removed by the tape. With the SiO_2 substrates there were some intermediate situations in which some portions of the gold film adhered and some portions were removed in an irradiated area. This made the determination of a threshold dose difficult. Furthermore, of the four samples used, one failed to show any adhesion after irradiation with a Cl beam at doses that produced enhanced adhesion on the other three samples. Nevertheless, we describe our results, such as they are, as our experience may be useful to other workers.

The clearest measurement was for the Ni beam. Only one threshold was observed, 2.3 $\times 10^{13}$ cm^{-2}, and all higher doses up to an order of magnitude above the threshold dose passed the Scotch-tape test. In the case of the Cl and Si beams, there was some evidence for two thresholds; for Cl at 2 and at 4 $\times 10^{14}$ cm^{-2}, and for Si at 4 $\times 10^{14}$ cm^{-2} and 2 $\times 10^{15}$ cm^{-2}. The lower thresholds define a power law of

$$D^{th} = 4 \times 10^{16} (dE/dx)^{-2.7 \pm 0.4} \ cm^{-2}$$

These results are similar to those reported in ref. 2. The upper thresholds are given by

$$D^{th} = 2 \times 10^{18} (dE/dx)^{-4.5 \pm 1.0} \ cm^{-2}$$

Two thresholds for the Scotch-tape test were observed by Wie, et al.[13] for the Au-SiO_2 system. The upper threshold observed by them for Cl bombardment corresponds to the lower threshold observed here.

The samples of fused silica exhibited macroscopic changes in the irradiated areas. The outline of the irradiated area was visible when viewed in oblique light, and cracks in the surface were easily observed with a binocular microscope. The former effect is a compaction of the SiO_2 that has been noted by Wie, et al.,[13] and by Berkowitz, et al.[14] in connection with bombardment for adhesion enhancement. The relatively large cracks we observe are probably not

directly related to the crazing of the surface seen at lower doses[13] and which disappeared above 4×10^{13} Cl ions $cm.^{-2}$

V. AUGER ANALYSIS OF THE AU-TA INTERFACE

Samples of the gold on tantalum system were analyzed with a scanning Auger microprobe at the Surface Science Laboratory of the Solid State Institute of the Technion-Israel Institute of Technology, Haifa.[15] Assuming that the native oxygen present at the interface is in the form of Ta_2O_5, the thickness of this oxide layer is about 40 Å. The samples that initially passed the Scotch-tape test without irradiation (as a result of sputter cleaning before evaporation) also had this same amount of native oxide. This is not unexpected since the argon gas used for sputtering was not of ultra-high purity and the base pressure of the evaporator system during sputtering was about 8×10^{-6} Torr. No differences at the interface were apparent from the Auger analysis that would suggest why the sputter-cleaned sample had a greater adhesion. The surfaces near the edges that had a level of adhesion sufficient to withstand abrasion showed traces of Cu, Zn and Sn at the interface. The small amounts of these elements were deposited during the sputter cleaning, having been removed from the brass sample holder. It is well known that Ni or Cu will increase the bonding of thermally deposited metallic thin films.

Figure 5a shows a depth profile for the relevant element concentration in an unirradiated region of a Au-Ta sample having a gold film 620 Å thick. The abscissa is the sputter-time, which is related to the depth in the sample. The only elements present at the interface with $Z > 3$ were gold, tantalum, and oxygen. Figure 5b shows the depth profile for oxygen in this region and an adjacent region that had been irradiated with an *actual* dose of 1.9×10^{14} cm^{-2} at an angle of 70°. (The 0° equivalent dose is 5.7×10^{14} cm^{-2}.) The main differences in Fig. 5b are the shift to larger sputter times and a distinct broadening of the oxygen profile in the irradiated region. The former is a result of the irradiated gold film having a sputter rate for 1.5 keV Ar ions about 13% slower than unirradiated gold.[15] The broadening of the oxygen profile cannot be explained solely by a change in the sputter rate but corresponds to a migration of oxygen

toward the tantalum side of the interface. After correcting for the decrease in the sputter rate, the oxygen profile in the irradiated region was still broader and skewed toward the tantalum side. Both of the oxygen peaks have the same area to within ten percent and thus do not indicate any gross change in the total amount of oxygen present at the interface before and after irradiation. Auger analysis of two other samples having 250 Å gold films and similar doses showed similar effects when comparing irradiated and unirradiated areas.

VI. DEPENDENCE OF THE ADHESION ENHANCEMENT ON THE LENGTH OF TIME FOLLOWING BOMBARDMENT

We often made a second peel test immediately after the first test. The Au-Ta samples yielded consistent results in these successive tests, i.e., irradiated areas that passed the first test also passed the second test. However, we discovered that some samples with 250 Å gold films on tantalum that initially passed a peel test did not pass when the test was repeated a week later. This is in contrast to the observatioL made for gold films on dielectrics, viz. that the strength of the bonding improved with time.[1] Since the potential applications of this method for adhesion enhancement depend on the permanence of the bonding, we investigated this time dependence further. Samples were prepared with gold films of 250 Å, 600 Å and 1600 Å, and with doses of 2, 4, and 8 $\times 10^{14}$ cm^{-2}. Peel tests at appropriately spaced time intervals were then performed. The results are summarized as follows:

The 250 Å samples exhibited a significant deterioration in adhesion during the first 36 hours. Areas irradiated at dosage levels twice the threshold (as measured just after bombardment) no longer passed the test. However, areas having four times the threshold dose still passed. After the initial 36–48 hours the rate at which gold was removed in successive tests slowed. A week after the bombardment only small amounts of gold from the edges of the surviving gold spots were removed by successive peel tests spaced five to ten days apart.

This removal of gold around the edges of an irradiated area was a consequence of a non-uniform dose. (The beam wobbler had not been used for these particular studies.) Indeed, the successive peel tests produced a qualitative profile of the dose over the irradiated area. A scratch made across the middle of the irradiated area did not result in any gold being removed at the edges of the scratch at later times. This demonstrated that the gold removed around the perimeter of the irradiated area was not a consequence of a substance diffusing along the interface from the edges of the gold spot.

The samples with 600 and 1600 Å films showed much less deterioration over the same time interval. For example, on a second test after 48 hours, areas irradiated with a dose 25% above the threshold dose still passed. (This is in contrast to the 250 Å film where a dose exceeding the threshold by at least a factor of two was required to pass the test after 36 hours). Four weeks after bombardment the areas that passed the second test at 48 hours continued to pass subsequent tests. Only around the edges of a spot would a minute amount of gold be removed. In order to measure the deterioration of adhesion enhancement more accurately, a sample was prepared with a 600 Å gold film and irradiated with doses increasing by a constant amount of 5×10^{13} cm^{-2} (0° equivalent). The dose required to pass the Scotch-tape test as a function of time is shown in Fig. 6. Thus, the rate at which the adhesion enhancement is lost decreases with time.

The loss of adhesion is reversible: reirradiating an untested portion of an area at a later time can restore the adhesion to the level achieved after the first bombardment. Samples of Au on Si showed a substantially slower loss of adhesion with time, even though the Au film was only 250 Å thick.

Samples that had been sputter-cleaned before the evaporation of the 250 Å of gold passed the Scotch-tape test without any irradiation (as noted earlier). These samples showed no deterioration of adhesion under repeated peel tests extending over two months. Thus, the

deterioration with time is associated with the adhesion enhancement that is induced by irradiating the interface.

VII. MISCELLANEOUS MEASUREMENTS AND OBSERVATIONS

A. The peel test

Two types of peel tests were used in these measurements. In the first type the tape was peeled from the surface by hand using a rapid pull in a direction normal to the surface. The only information in this measurement is the threshold dose, which is the minimum dose required to make the thin film adhere to the substrate rather than to the tape. In the second type of measurement the weight required to peel a narrow strip of tape (2-3 mm wide) was measured. This measurement provides more information, in that one can measure the peel strength (g/mm) required to peel the gold film from the substrate. This quantity, p, is of primary interest and should vary continuously with the dose. When the gold film adheres to the substrate, however, it is the peel strength to remove the tape from a gold surface that is observed. This value is only a lower limit to p. This second type of measurement, which we shall refer to as the continuous test, is similar in principle to the method used by Baglin et al.[7] although our technique is not as sophisticated . In our case the force was measured using an ordinary laboratory balance of the type suitable for weighing quantities of 0.1 to 1000 gm, and the change in dose was discrete rather than continuous. The continuous peel test enabled the following observations:

1. The continuous peel strength for Scotch-tape on unirradiated surfaces:

The average peel strength for Scotch-tape on an unirradiated solid gold or tantalum oxide substrate cleaned in ethanol was about 10 g/mm. Individual measurements with different strips of tape of varying widths gave results within 10% of the average value. Furthermore, the peel strength was the same within 20% for repeated tests done on the same surface but without inter-

mediate cleaning. This fact is important for the measurements in which the peel test was repeated at later times on the same location of an irradiated thin film.

Evaporated thin gold films that adhered to the tantalum substrate in a Scotch-tape test without prior irradiation had higher peel strengths, of about 15-16 g/mm.

2. The effect of radiation on the peel strength for the *tape-to-gold* interface:

Measurements of the peel strength for Scotch tape on various gold surfaces were made. The peel strengths are shown in Fig. 7 as a function of the actual dose. An increase of peel strength with dose is observed with a solid gold substrate irradiated by Cl ions for doses below $3 \times 10^{13} cm^{-2}$. Above this value the peel strength saturates to a value of roughly 21 g/mm. This observation suggests that the change in peel strength arises from the accumulation of hydrocarbon molecules on the surface of the gold. (A dependence of peel strength on dose was observed in ref. 7 for Scotch tape on solid copper. However, the effect was in the opposite direction!)

The tape-to-gold peel strength for various Au-Ta and Au-Si samples irradiated with Si, Cl and Ni beams prepared with the usual glow-discharge cleaning could only be measured at doses equal to or larger than the threshold dose. It was observed to be constant above the threshold dose. Fig. 7 shows the value of the peel strength at threshold. We observe that the peel strengths for the tape-gold interface are all similar for actual doses at or above 2×10^{13} cm^{-2}, and that the saturation peel strength for the irradiated solid gold strip is similar to the value for the irradiated thin films once the actual dose exceeds 2×10^{13} cm^{-2}. The average peel strength is 21 g/mm, with standard deviation 3 g/mm. A layer of hydrocarbon molecules on the surface of the irradiated thin films was identified in the Auger analysis, supporting the interpretation that the change in peel strength with dose is due to the accumulation of a hydrocarbon layer. The dose required to achieve the saturation value of the tape-to-gold peel strength is about the same as the smallest threshold dose encountered. It follows that the increase in tape-to-gold

peel strength associated with irradiating the surface does not significantly affect the relative values of the threshold doses required to pass the Scotch-tape test.

Another observation also supports the above conclusion. The threshold doses required at angles of incidence of 0° and 85° differ by an order of magnitude. However, both the Scotch-tape peel test and the cotton bud abrasion test give the same results (i.e., a cos θ dependence).

3. Continuous peel test at doses below threshold:

Several measurements were made of the peel strength for doses below the Scotch tape threshold, i.e., for doses where the tape peels the gold film from the substrate. Unirradiated areas had peel strengths of typically 0.5 g/mm (i.e., a force/length of 5000 dyne/cm). This same strength would also remove the gold from the substrate for doses up to one half to three-quarters of the threshold dose. Above this point the peel strength would rise very rapidly with increasing dose until the threshold dose was reached, at which point the gold no longer adhered to the tape. At threshold the peel strength was typically 21 g/mm, which is equivalent to a force of 2×10^5dyne/cm. A second observation is that the absolute values of the threshold peel strength are several orders of magnitude *larger* than an upper limit obtained by equating the energy of adhesion to the energy of cohesion for gold or tantalum. This may be seen as follows: The tensile strength, τ, of gold is 1.3×10^9 dyne/cm. This is the force per unit area required to separate a gold-gold interface. If we assume this force acts over a distance d, then the energy of cohesion, 2γ, is given by $2\gamma = \tau d$. An upper limit[16)] for d is 10 Å, and thus the energy of cohesion for Au is $2\gamma \lesssim 1.3 \times 10^2$erg/cm^2. The average force per unit width to peel the gold film was 2×10^5 dyne/cm which corresponds to an energy of adhesion of 2×10^5 erg/cm^2.

These above characteristics of the peel test probably arise from the visco-elastic properties of the tape and polymer adherate. As the tape is 10^4 times as thick as the gold foil, the visco-elasticity of the tape makes an important contribution to the peel strength. It is thus practically

impossible to measure an absolute value for the energy of adhesion of the Au-Ta interface by this technique.

4. Reproducibility of the threshold peel test:

Separate peel tests were made using irradiations on different areas of the *same* Au-Ta sample. Comparison of the deduced threshold doses in these cases permitted an assessment of the reproducibility of the Scotch-tape test. We conclude from four such comparisons that the reproducibility is ±10%. If we consider the results for threshold peel strengths obtained with different beams and different samples (Fig. 7) then the peel strengths appear reliable to ±15%. Therefore, independent of the absolute peel strength required to remove the tape, the *threshold dose* is found to be a sufficiently reproducible quantity to be a useful indicator of adhesion enhancement.

B. Dependence of the threshold dose on the thickness of the gold film

A Au-Ta sample with a gold film of 250 Å on one-half the area and 500 Å on the other half was prepared in a single evaporation without breaking the vacuum. The threshold dose for the 500 Å film was 40% higher than for the 250 Å film. A less precise comparison can be made by considering the typical threshold doses for 250 Å and for 500-600 Å films on different samples. This indicates that the threshold for the thicker films (on Ta) is about twice that of the 250 Å film. We conclude that the threshold dose, as measured by the Scotch-tape test, does depend on the thickness of the film. This may arise from the nature of the test itself. Possibly the thickness of the gold film may affect other properties (such as the density of micro-cracks in the gold) that determine the threshold for peeling.

C. Wettability of the irradiated surface:

The bombardment of the surface and interface of a sample has a number of consequences. For example, it was found in ref. 2 that the irradiated interface, exposed by removal of the gold

film, would be preferentially wet by alcohol. Related phenomena were observed here and are as follows:

The irradiated areas of the Au-Ta and Au-Si samples were nearly always visible to the naked eye after the bombardment. This may be connected to some extent with the deposition of hydrocarbons on the irradiated surface. In the few cases where the beam spot was difficult to see, it became visible when the sample was exposed to water vapor (the "breath test").

When the gold film was removed by the tape, the areas of irradiation could not be discerned at all with the naked eye. The breath test, however, easily revealed all irradiated areas, even for the smallest doses. This effect cannot arise from hydrocarbon deposition during irradiation, as these hydrocarbons exist only on the surface of the gold film and are therefore removed with the film. This enhanced wettability persisted with time. Cleaning the Ta substrate in nitric acid and in alcohol did not remove the effect. It may be connected to the modification of the interface that was revealed by the Auger analysis, which showed migration of oxygen in the region of the interface.

VIII. DISCUSSION

In sections A-C we first discuss the following three results:

(i) the very different behavior of the Au-Ta systems studied here and in refs. 2 and 4,

(ii) the decrease of adhesion with time, and

(iii) the migration of oxygen at the interface as a result of bombardment with high energy ions.

When these results are considered in light of experiments by S.V. Pepper[17] (on how adsorbed gases can affect the adhesion of a metal to an aluminum oxide substrate, section D), two possible mechanisms for adhesion enhancement become apparent (section E). After discussing these mechanisms, the significances of the angular dependence and the dE/dx dependence of the threshold dose are considered in sections F and G.

A. The Au-Ta results

The Au-Ta systems studied here exhibit threshold doses for the Scotch-tape test that are very different from those studied in refs. 2 and 4. The specific origin of these differences is not known. It seems most probable that the tantalum samples used in ref. 2 had a native oxide layer sufficiently thick (as did our samples) that the adhesion occurs at the interface of the gold film and a tantalum oxide substrate. If this is true, then differences between the sets of samples probably originate with differences in the preparation of the surface. One factor to consider is the glow-discharge cleaning, which was used in the present work but not in ref. 2. In this procedure the samples remained in the evaporator for several hours after the glow-discharge while the system pumped down to a pressure less than 10^{-5} Torr. Hence, adsorption of ambient gases occurred before the evaporation was performed.

The glow discharge cleaning by itself, however, could not be responsible for the entire difference between the earlier and the present results. Although a sample prepared without the glow-discharge cleaning had a somewhat lower threshold dose than similar samples prepared with the discharge ($0.7^{+0.2}_{-0.1} \times 10^{14}$ cm^{-2} versus 1-2 $\times 10^{14}$ cm^{-2}) it nevertheless required a dose three times larger than anticipated from the results of ref. 2.

It is possible that the origin of the difference in the results rests with trace amounts of gases adsorbed on the substrate at the time the gold film was deposited. With two exceptions, the Auger analysis limits the presence of such elements to a few percent of an atomic layer. The two potentially important exceptions are oxygen (since the substrate is tantalum oxide) and hydrogen. What seems remarkable is that the presence of adsorbed gases would affect not only the threshold dose, but also the power-law dependence of the threshold dose on the stopping power of the ion, i.e., $(dE/dx)^{-1.6}$ in ref. 2 and $(dE/dx)^{-3.0}$ here.

B. The time dependence

The adhesion enhancement decreases with time following the bombardment. The rate at which the adhesion deteriorates depends on the thickness of the gold film and thus is not an intrinsic property of the bonds formed at the interface as would be, for example, the formation of a metastable bond that decayed with a characteristic mean lifetime. The form of the time dependence (Fig. 6) suggests a diffusion-controlled process since it resembles the dashed line, which is of the form $a + b\sqrt{t}$. This implies that adhesion enhancement can be affected by the introduction at the interface of atoms that were not present at the conclusion of the bombardment.

C. The migration of oxygen

The observed migration of oxygen toward the tantalum side of the interface indicates that energetic ions interacting mainly through electronic energy loss mechanisms can cause a displacement of atoms at the interface. (A displacement or nonregistration of silicon atoms at a Au-SiO_2/Si interface can be induced by bombardment with energetic oxygen ions.[6]) While these displacements and migration may be caused by a variety of mechanisms, the important point is that movement does occur and (as suggested by the discussion in sections A. and B. above) the introduction or removal of very small quantities of atoms at the interface may significantly affect adhesion. This latter possibility is demonstrated clearly in the results of the experiments described in ref. 17.

D. The effect of adsorbed gases on metal-oxide shear strength

In a series of elegant experiments, S.V. Pepper has shown that amounts of adsorbed gases, in quantities less than a monolayer, can have a dramatic effect on the shear strength of a metal ball in contact with an Al_2O_3 (sapphire) surface.[17] He measured the force required to move a metal ball of, e.g., gold, copper or nickel, resting with a known perpendicular force on a flat sapphire crystal in an ultra high vacuum environment. Controlled amounts of gases such as oxy-

gen, nitrogen, or chlorine could be adsorbed onto the metal surface. In this way, it was shown that a fraction of a monolayer chemisorbed on the surface of the metal could strongly affect the static coefficient of friction between the metal and oxide surfaces. Oxygen and nitrogen enhanced the shear strength while chlorine caused it to decrease. In cases where a gas would not chemisorb onto the metal (oxygen onto gold) before it was placed in contact with the sapphire, activation of the gas by bombardment with 150 eV electrons produced chemisorption and a subsequent increase in shear strength.

E. Possible mechanisms for the adhesion enhancement

The above observations on shear strength together with the results of the present experiments suggest two possible interpretations for the adhesion enhancement arising from irradiation of the interface.

1. Oxygen adsorbed onto the Ta_2O_5 substrate at the time the gold film is deposited is subsequently activated by the passage of radiation through the interface and becomes "chemisorbed" to the gold film. After bombardment, oxygen diffusing through the gold film to the interface replaces (or deactivates) some of the activated oxygen and reduces the adhesion. Bombarding the surface a second time would then restore the adhesion enhancement, as was observed.

2. Atoms at the interface, whose presence weakens the adhesion, are dispersed as a result of bombardment, thus enhancing adhesion. This dispersion might occur through diffusion along radiation-induced defects in the film or substrate or because of local, microscopic heating. After bombardment these atoms diffuse back to the interface (or other atoms diffuse through the gold film to the interface) with a corresponding reduction in adhesion.

These two possible mechanisms are not mutually exclusive, nor are they expected to be the only possible explanations. However, the importance of trace amounts of adsorbed atoms at the interface on adhesion enhancement is suggested by these studies and it seems to correlate

a number of observations. This conclusion has also been made by Baglin.[8)]

F. The dependence of the threshold dose on angle

This measurement was made in order to investigate geometrical properties of the interface where the adhesion enhancement takes place. Such properties are the thickness of the region (normal to the plane of the interface) over which bonding takes place, and the area in the plane of the interface affected by the passage of an ion through the surface. Since the measurement of dose dependence on the angle of the penetrating ion with respect to the interface is geometrical in nature, one expects resulting information on the interface to be similar in kind.

Figure 8 illustrates four different geometrical combinations for characterising the ion and the interface. Figure 8a is the extreme (and unrealistic) case of an interface having no thickness and an ion path represented by a line. For angles less than 90° the intersection of a line and a plane is a point and this intersection is independent of angle. In this case the threshold dose would be a constant. This is not observed, as may be seen in Figs. 3 and 9. In the latter figure the results are presented as the ratio of the threshold number of particles through a collimator fixed normal to the beam for an interface angle of θ with respect to the beam, $N_p^{th}(\theta)$, and the corresponding number at normal incidence, $\theta = 0$. (This ratio is proportional to the dose in Fig. 3 divided by cos (θ).) The predictions obtained under the various geometrical assumptions in Fig. 8 are shown in Fig. 9.

If the ion's influence in producing bonds extends for a distance R normal to its direction of motion, then the number of bonds it can be expected to form is proportional to the area of intersection of a cylinder of radius R with a plane (see Fig. 8b). This area is proportional to $(\cos\theta)^{-1}$, and one would expect the dose to vary as cos θ. The experimental results are consistent with this.

Figure 8c depicts a case in which the perpendicular distance over which bonding occurs is finite. In this case one may ask whether it is just the stopping power, dE/dx, or the total energy lost in the interface region, $\left(\frac{dE}{dx}\right) \cdot \Delta x$, which determines the threshold dose. In the latter

case, an observed stopping-power dependence of $\left(\frac{dE}{dx} \bullet \Delta x\right)^{-1.6}$ at normal incidence would imply an angular dependence of $(\cos\theta)^{1.6}$ since Δx is proportional to $(\cos\theta)^{-1}$. The experimental results are clearly inconsistent with this, and would have an even greater discrepancy with the prediction $D^{th} \propto (\cos\theta)^{3.0}$ based on the $\frac{dE}{dx}$ dependence observed here. This result indicates that the average energy lost per atom is the relevant quantity even if the interface has a finite thickness.

The remaining possibility, indicated in Fig. 8d, combines the elements of a cylinder of influence about the ion's path and an interface of finite thickness. In considering this we make the assumption (a) that a bond occurs only when all the atoms in a line perpendicular to the interface are included in the volume swept out by the ion's path. In this case changing the angle of the ion has two effects that work in opposite directions: in a deviation from normal incidence the areas of intersection with the upper and lower planes defining the interface increase, but the mutual overlap of these two areas in the normal direction, required by assumption (a) above, decreases. The detailed dependence of the dose on the angle depends on the ratio of the half thickness t of the interface and the radius of influence, R, of the ion, $D^{th} \propto \frac{\cos\theta}{1 - \frac{t}{r}\sin\theta}$. Predictions for various values of t/R are shown in Fig. 9. Given this geometrical interpretation and its assumptions, our results indicate that the radius of influence of the ion is at least 5 times greater than the half thickness of the interface.

G. The dependence of the threshold dose on the bombarding ion

Until the present work the only systematic study of adhesion enhancement on the type of the bombarding ion was that of ref. 2. There it was found that the electronic stopping power, dE/dx, was a suitable quantity for parametrizing the results, and specifically that $D^{th} \propto \left(\frac{dE}{dx}\right)^n$ where n = -1.6. Here we observe that, while a power law for the threshold dependence on dE/dx remains a suitable parametrization, the value of n can vary significantly from case to case. We observe values of -3.0 ± 0.2 (Au - Ta_2O_5/Ta), ≈ -3 (Au - SiO_2), and -4.1 ± 0.3 (Au - SiO_2/Si). Since any model for the adhesion enhancement mechanism will have to explain the relative effectiveness of different ions in producing adhesion, we discuss here the significance of the observed non-proportional behavior of Dth on dE/dx (i.e., $n \neq -1$).

For a given stopping medium the stopping power of an ion depends on its atomic number and on its velocity. We have deliberately chosen the energies of the different ions used here to correspond to the same velocity. Thus the cross sections for the different modes of electronic energy loss (valence electron ionization, inner core electron excitation, etc.) will be the same in first order except for a scale factor Z_{eff}^2 where Z_{eff} is the equilibrium charge of the ion at velocity v in the stopping medium.[18] Thus, $\frac{dE}{dx}$ also varies approximately as Z_{eff}^2, and the number of delta rays, secondary and tertiary electrons produced per unit volume should have the same proportionality. (We are neglecting saturation effects.) If we calculate in this picture the average number of electrons per unit (electron) energy crossing the interface as a result of the passage of one ion, the result will be proportional to dE/dx. If the adhesion enhancement represents bonds being formed as the result of activation by electrons crossing the interface, then we might expect the number of bonds to be proportional to the number of electrons and the threshold dose to scale as $(dE/dx)^{-1}$. Since this scaling is not observed, one must either consider models in which the number of electrons crossing a surface is not proportional to dE/dx or look for adhesion mechanisms involving collective, non-linear effects. An example of the former is the assumption of local thermalization around the ion's path and a Fermi-gas

distribution of electron energies—the Dushman-Richardson equation for thermionic emission.[4] Examples of collective, non-linear mechanisms are found in attempts to correlate the adhesion enhancement with track formation in insulators or with sputter yields.[1]

1. The Dushman-Richardson equation:

Derived from the free electron theory of metals, the following equation describes the number of electrons N ($cm^{-2}s^{-1}$) crossing an interface with a work function ϕ for a system in equilibrium at temperature T.

$$N \propto T^2 e^{-\phi/kT} \tag{1}$$

In order to calculate the local temperature from the stopping power of the ion we use the specific heat for a free electron Fermi gas,

$$\frac{d\langle E\rangle}{dT} = \pi^2 k^2 \frac{T}{2\,E_F} \tag{2}$$

where E_F is the electron Fermi energy.

This yields, after integration,

$$T \propto \sqrt{\frac{dE}{dx}} \tag{3}$$

The constant of proportionality involves the volume over which local thermal equilibrium is assumed to occur.

Assuming $D^{th} \propto N^{-1}$ and combining equations (1) and (3), the fits to the data shown in Fig. 10 are obtained. These fits involve two parameters, an overall normalization and a parameter ϕD representing the product of the work function and the dimension of the equilibrated volume. The latter is assumed to be a cylinder normal to the interface with a radius of D atomic diameters.

The following values of ϕD are obtained: for $(dE/dx)^{-1.6}$, $\phi D = 36$ eV: for $(dE/dx)^{-3.0}$, $\phi D = 140$ eV: and for $(dE/dx)^{-4.1}$, $\phi D = 300$ eV. A value of $\phi D = 36$ eV is equivalent to a work

function of 3.6 ev and an equilibrated volume involving ~300 atoms per atomic layer. At dE/dx = 500 ev/atomic layer, this corresponds to a local temperature of 2 x 10^4 °K. If one accepts the strong assumptions in this model (e.g., attainment of local thermal equilibrium) then a value of 10^4 °K may not seem farfetched.

2. Collective mechanisms:

Track formation in insulators and sputtering are examples of collective effects. In the former case, adjacent atoms must be sufficiently ionized for a mutual Coulomb repulsion to overcome the restoring force of the lattice. The important quantity determining this is the primary ionization rate[9] $\frac{dJ}{dx}$. The energy contained in mutual Coulomb repulsion is proportional to $\left(\frac{dJ}{dx}\right)^2$. If the mechanisms for track formation and adhesion enhancement were related, one might expect that the threshold dose would vary as $\left(\frac{dJ}{dx}\right)^{-2}$, since $\frac{dJ}{dx}$ and $\frac{dE}{dx}$ have very similar dependencies on ion velocity and atomic number, $\left(\frac{dJ}{dx}\right)^2 \propto \left(\frac{dE}{dx}\right)^{\approx 2}$. For the ions used in the present study, $\left(\frac{dJ}{dx}\right)^2 \propto \left(\frac{dE}{dx}\right)^{2.3}$.

At low bombarding energies the variation of sputtering yield (of UF_4, H_2O, and SO_2) with bombarding energy has been parametrized[10] as proportional to $\left(\frac{dJ}{dx}\right)^4$. Were the adhesion enhancement related to the sputtering mechanism, then we might expect a threshold dose varying as $\propto \left(\frac{dE}{dx}\right)^{-4.6}$. On the other hand, the sputtering of Al_2O_3 and Li Nb O_3 by Cl ions follows a dependence well described[19] by $(dE/dx)^{1.0}$.

It seems likely that, as different film-substrate combinations are studied in the future, a range of exponents will be observed. Thus, a model's agreement with a particular exponent for a particular system will not be sufficient to establish a mechanism. Other information must be included as well. When this is done, the case for the above collective mechanisms is consider-

ably weakened. For example, as mentioned above, the sputtering yield for high energy chlorine ions seems to scale nearly linearly with dE/dx, (ref. 20), and there does not appear to be a strong correlation of the threshold value of dE/dx for track formation with the threshold dose for adhesion enhancement.

Thus, the origin of the power law dependence of the threshold dose on dE/dx, and the values of the exponents for different systems remain an open and intriguing question for further study.

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Table I.

Threshold doses for the Scotch-tape test. 500-600 Å of gold on a tantalum substrate (with ~40 Å of native oxide), irradiated by beams at 2.85 MeV/a.m.u.

Beam	dE/dx[a)] (MeV cm^2/mg)	D^{th} (0° equivalent)[b)] (cm^{-2})
$^{12}C^{5+}$	1.28	$(5.1 \pm 2.0) \times 10^{16}$
$^{16}O^{5+}$	2.11	$(1.0 \pm 0.3) \times 10^{16}$
$^{28}Si^{7+}$	4.86	$(1.5 \pm 0.3) \times 10^{15}$
$^{35}Cl^{8+}$	6.36	$(3.3 \pm 0.7) \times 10^{14}$
$^{58}Ni^{12+}$	11.90	$(6.4 \pm 1.6) \times 10^{13}$

[a)]For ions stopping in Au. From ref. 12.

[b)]The angle between the beam and the normal of the sample was 70°.

Table II.

Threshold doses for the Scotch-tape test. 250 Å of gold on a silicon substrate (with native oxide), irradiated by beams at 2.85 MeV/a.m.u.

Beam	dE/dx[a)] (MeV cm^2/mg)	D^{th} (0° equivalent)[b)] (cm^{-2})
$^{28}Si^{7+}$	4.86	$(9.7 \pm 2.4) \times 10^{15}$
$^{35}Cl^{8+}$	6.36	$(2.5 \pm 0.5) \times 10^{15}$
$^{28}Ni^{12+}$	11.90	$(2.5 \pm 0.5) \times 10^{14}$

[a)]For ions stopping in Au. From ref. 12.

[b)]The angle between the beam and the normal of the sample was 60°, 60°, and 80° for beams of Si, Cl, and Ni, respectively.

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Figure Captions

Fig. 1. Schematic diagram of the vacuum chamber in which the samples were irradiated.

Fig. 2. Photograph of a Au-Ta sample after irradiation and application of the Scotch-tape test. The dose increased stepwise from left to right. The circles of gold remaining on the substrate are from bombardments at 0° (normal incidence) and the ellipsoidal-shaped areas of gold were irradiated at 80°. Adjacent circles and ellipsoidal areas were exposed to equal numbers of particles. A slight misalignment of the sample is responsible for the changing location of the ellipsoids relative to the circles. The row of circles at the top of the sample represents a series of bombardments in coarse steps (factors of $\sqrt{2}$) to locate the threshold. The second series of bombardments at 0° and 80° was made using finer steps near the threshold dose.

Fig. 3. The threshold dose required to pass the Scotch-tape test as a function of angle, normalized to normal incidence.

Fig. 4. The threshold dose (at normal incidence) for different bombarding ions, as a function of the ion's differential energy loss in gold. In order of increasing dE/dx, the open squares correspond to ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{58}Ni beams, all at 2.85 MeV/nucleon. Note the difference in both magnitude and slope of the results for Au-Ta systems obtained here and in ref. 4. The straight lines corresponds to $(dE/dx)^{-1.6}$ for the dashed line, $(dE/dx)^{-3.0}$ for Au-Ta, and $(dE/dx)^{-4.1}$ for Au-Si.

Fig. 5. Scanning Auger Microprobe analysis of Au-Ta samples: a) before irradiation. b) a comparison of oxygen depth profiles at adjacent locations on the same sample, one of which had been irradiated with 1.9×10^{14} ions per cm^2 at an angle of 70°.

Fig. 6. The threshold dose required for a spot on a 600 Å Au-Ta sample to pass the Scotch-tape test if the test is applied a period of time after the bombardment. The increased dose required as a function of time after the bombardment corresponds to

a loss of adhesion with time. The dashed line is of the form $D^{th} = a + b\sqrt{t}$ where a and b are adjusted constants and t is the time after bombardment.

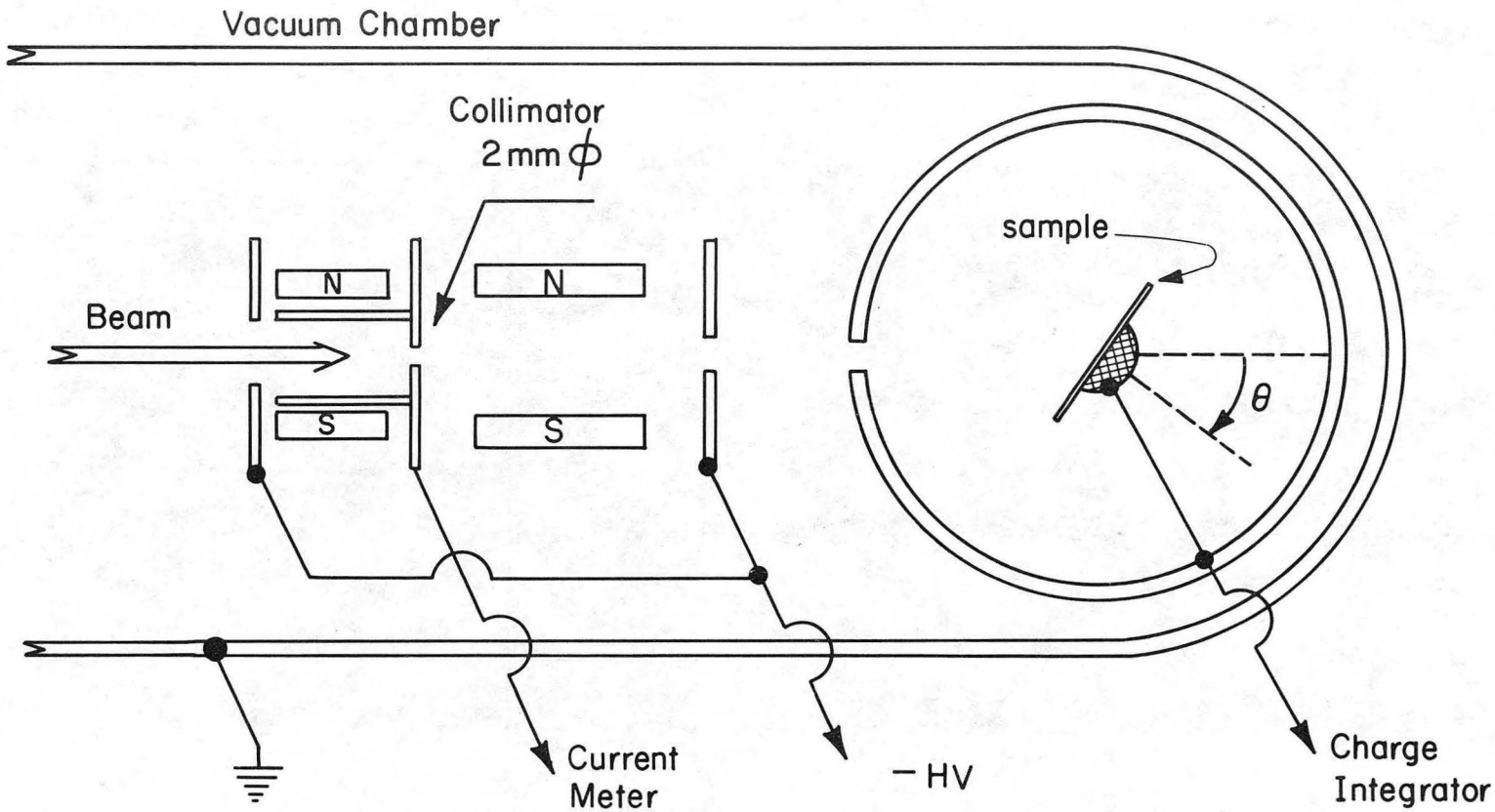
Fig. 7. Peel strengths to separate the tape from the gold surface, measured as a function of actual dose. The force needed to separate the tape (peeled at 90° from a gold surface), in dynes per centimeter width of the tape, is equal to the ordinate multiplied by 9800. The peel strengths for an unirradiated solid piece of gold and an evaporated gold film are different and are indicated. The peel strength on an irradiated surface of solid gold increases with dose (solid points). Peel strengths for various samples of a thin gold film on a substrate, irradiated by different beams (circles), are given as a function of dose.

Fig. 8. Purely geometrical representations of the interface and the trajectory of an ion crossing it. a) The interface is a plane having no thickness and the ion's trajectory is a line. The intersection is a point. b) The ion's influence in causing additional bonding extends for a distance R perpendicular to its central ray. c) The region over which interfacial bonding occurs has a finite thickness, 2t. d) Both interface and ion trajectory have finite extent. e) For the case of the geometry shown in d), the large ellipses represent the intersection of the ion trajectory with the upper and lower planes, defining the extent of the interface. The hatched areas are the part of the volume that is within the ion's trajectory and that connects vertically to both upper and lower planes.

Fig. 9. The results given in Fig. 3, divided by cos θ. The ordinate is written as the ratio of the number of particles passed through the beam collimator, at the Scotch-tape threshold, for the sample tilted at angle θ and at normal incidence. The solid lines are the predictions for the various geometrical possibilities shown in Fig. 8.

Fig. 10. Calculations of the threshold dose using the Dushman-Richardson equation (1) to predict the number of electrons crossing an interface having a work function ϕ.

Local thermal equilibrium is assumed to exist for a radius of D atomic diameters perpendicular to the ion's path. The slope of the curve is determined by the parameter ϕD, which has the indicated value.



XBL 855-2632

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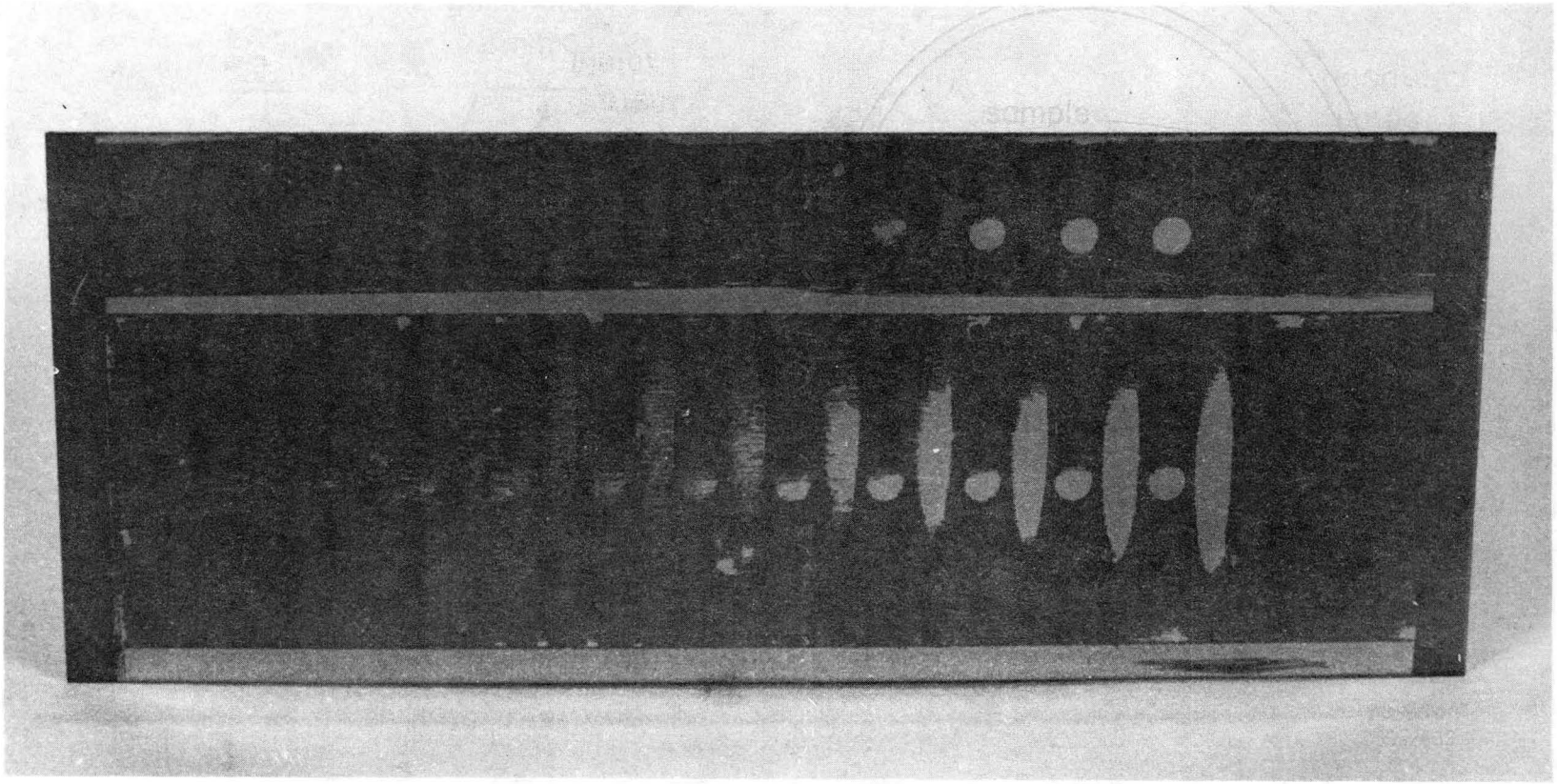

Fig. 2

XBB 855-4232

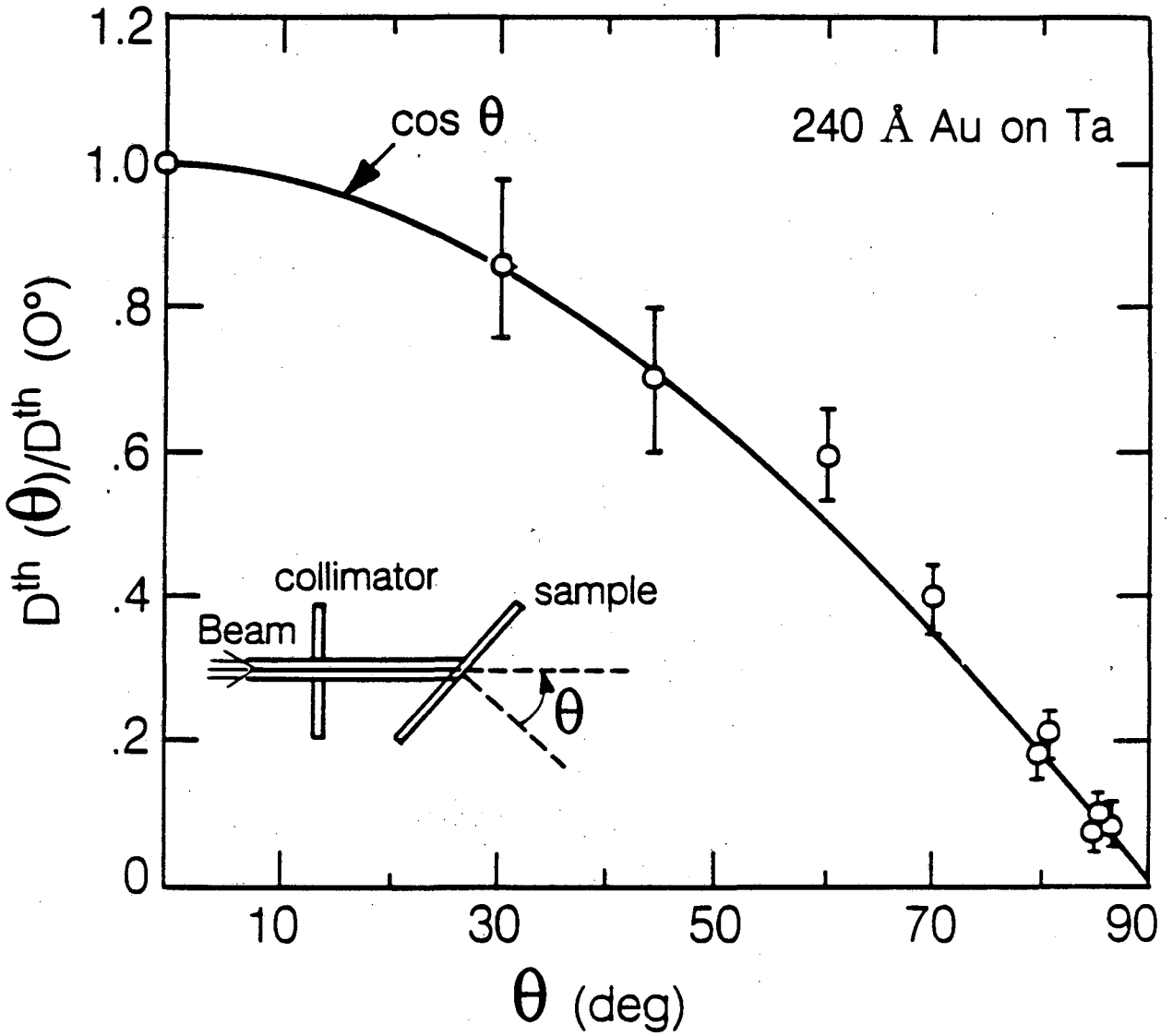

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Fig. 3.

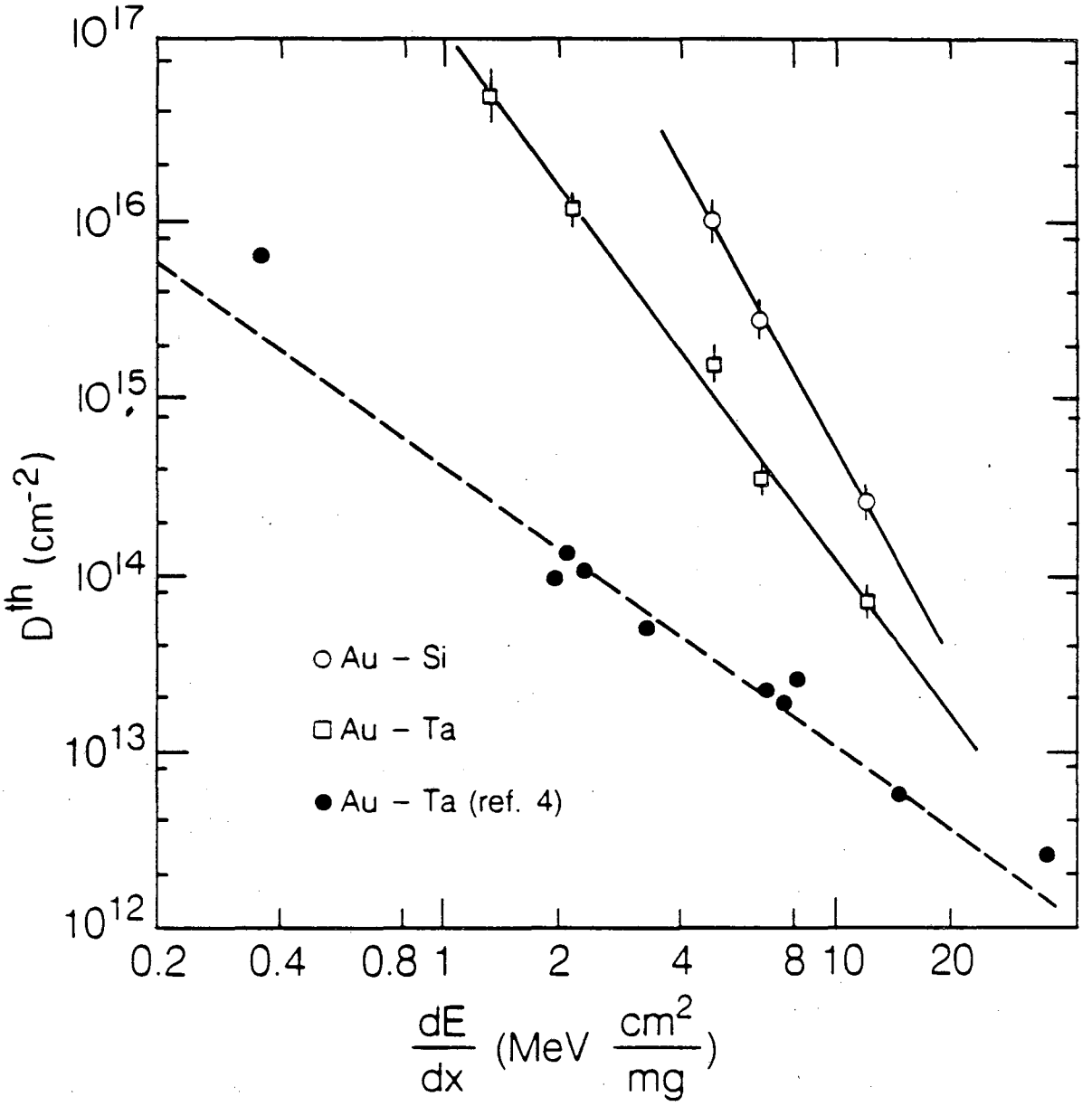

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Fig. 4.

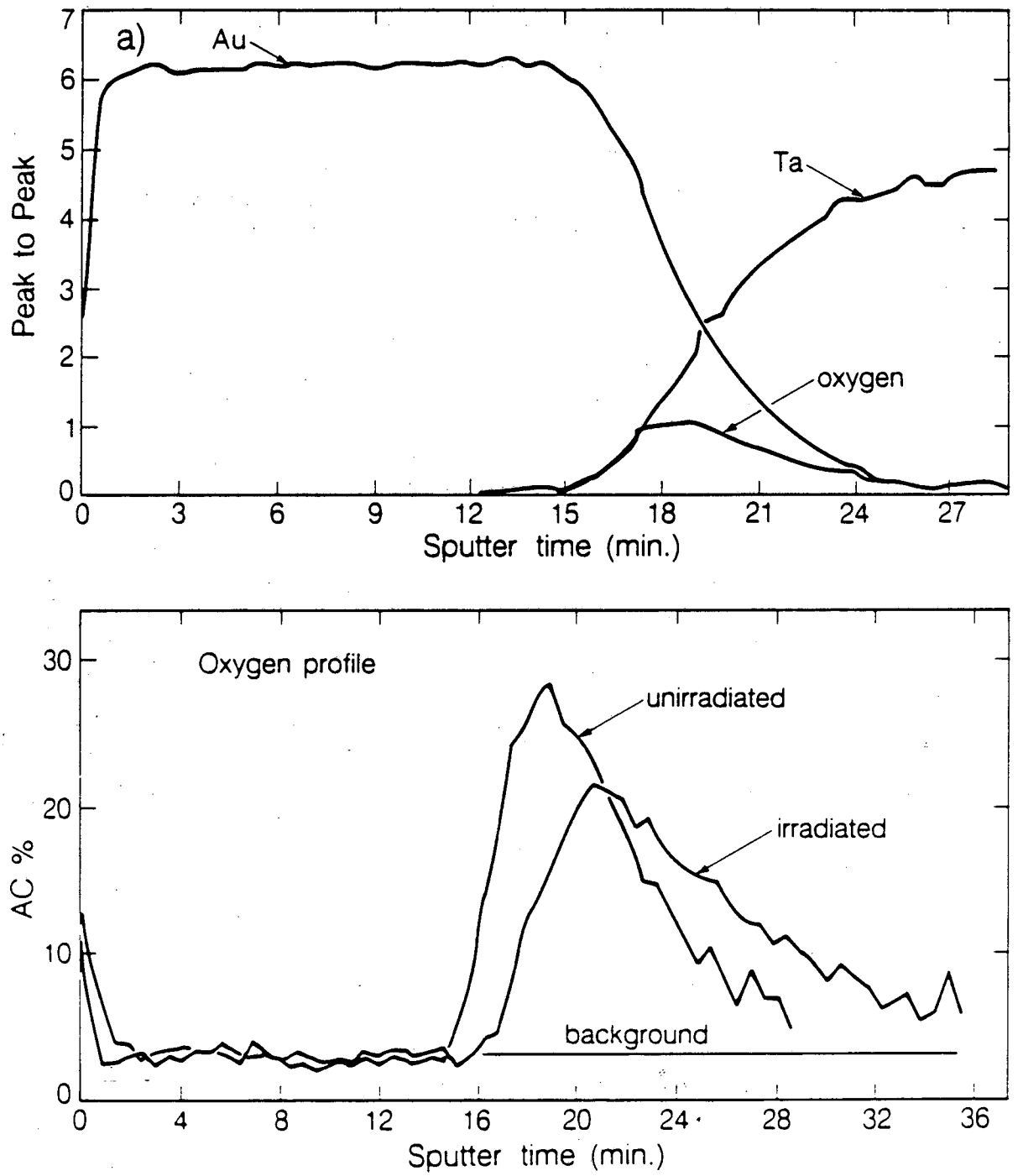
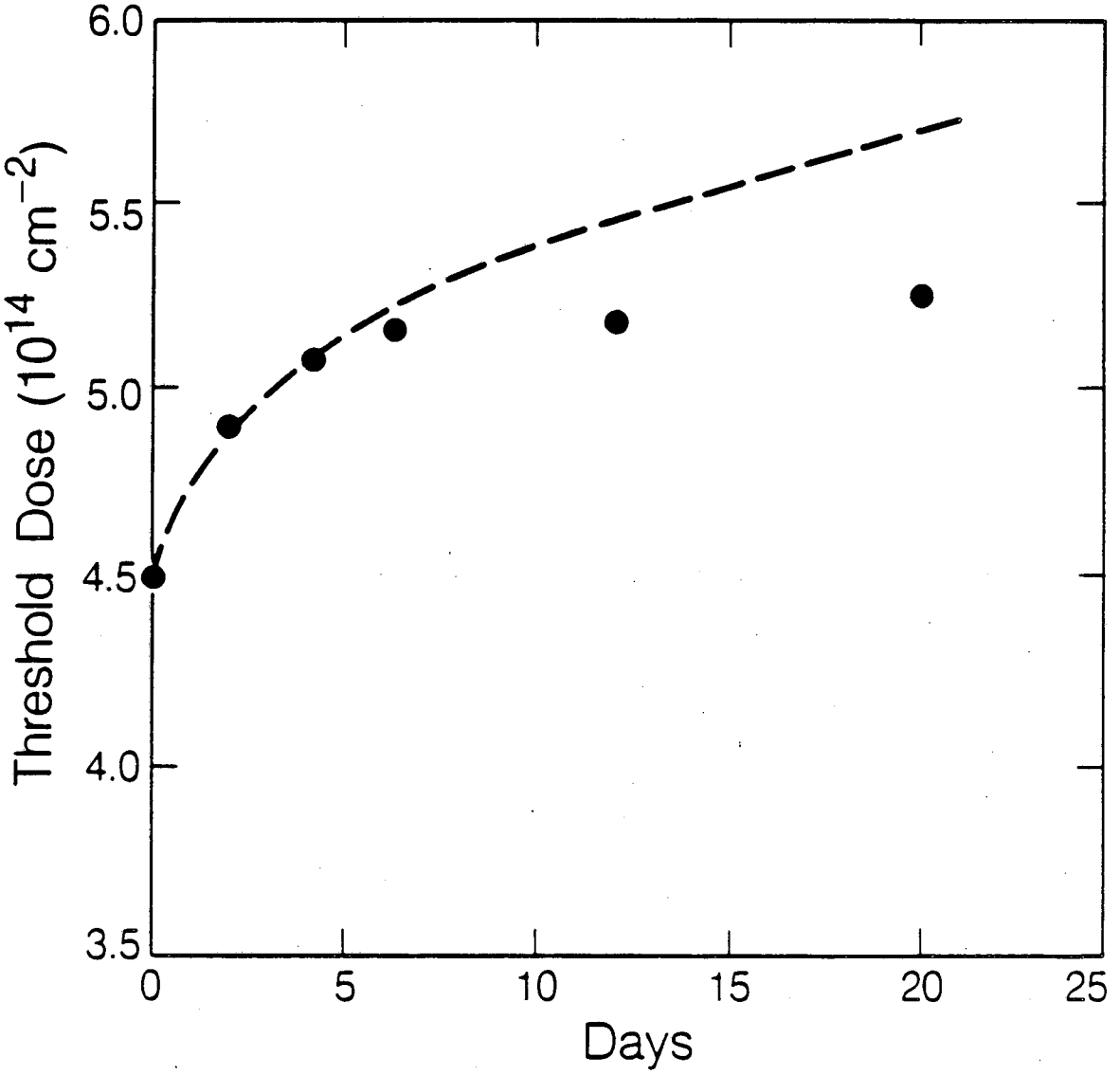

a)
Au
Ta
oxygen
Peak to Peak
Sputter time (min.)
Oxygen profile
unirradiated
irradiated
background
AC %
Sputter time (min.)
XBL 855-9977

Fig. 5.



XBL 855-9975

Fig. 6.

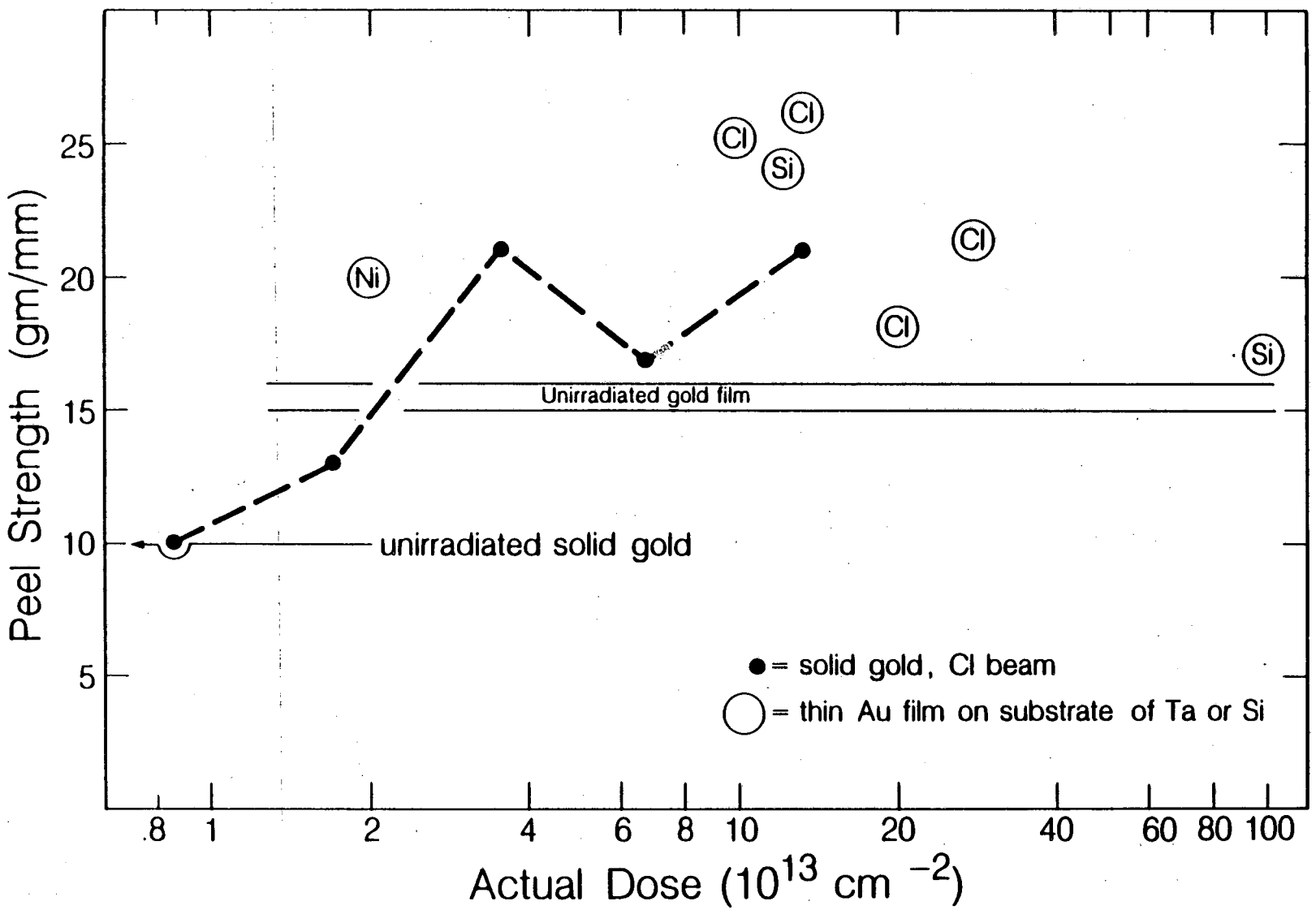


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Fig. 7.

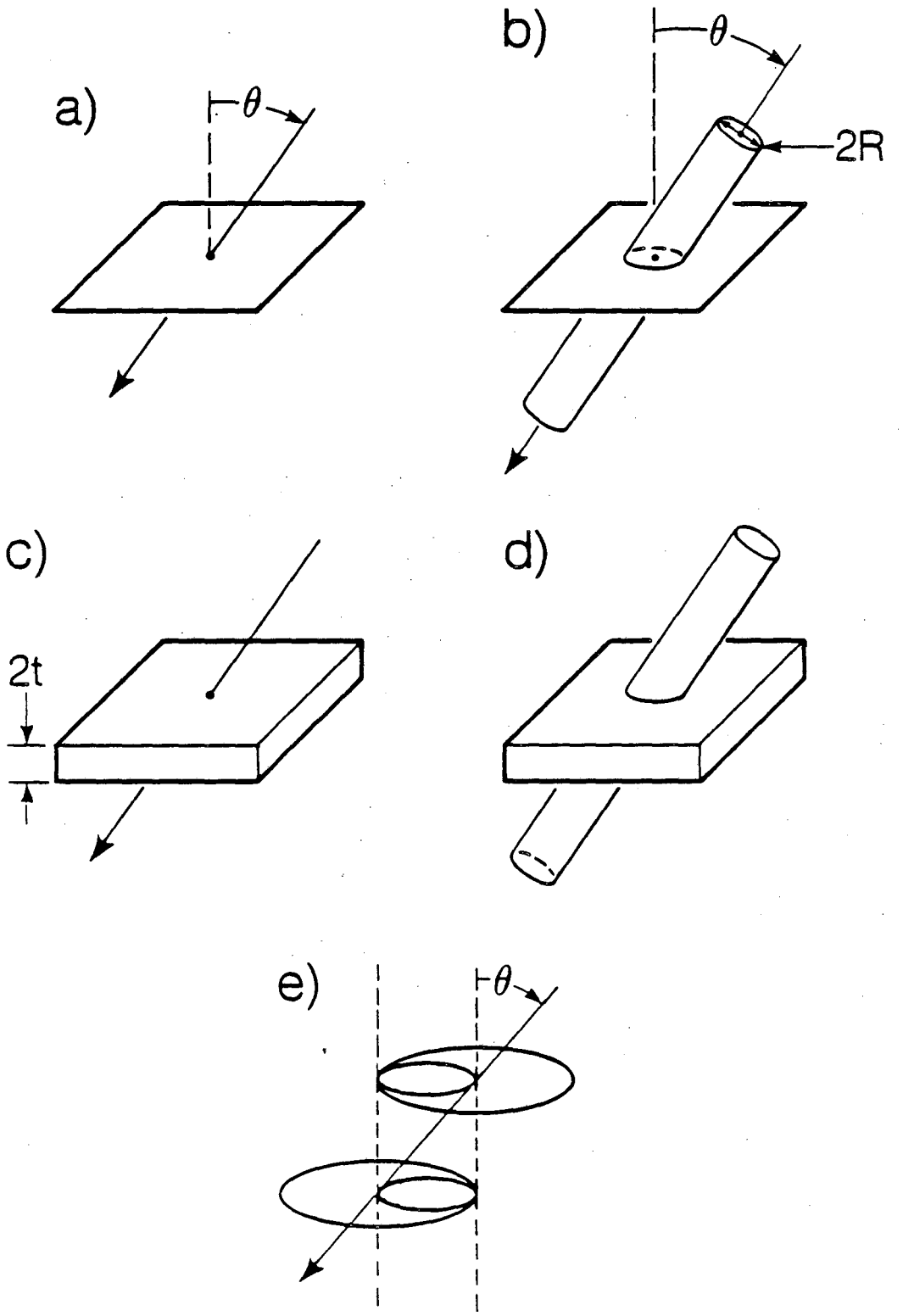

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Fig. 8.

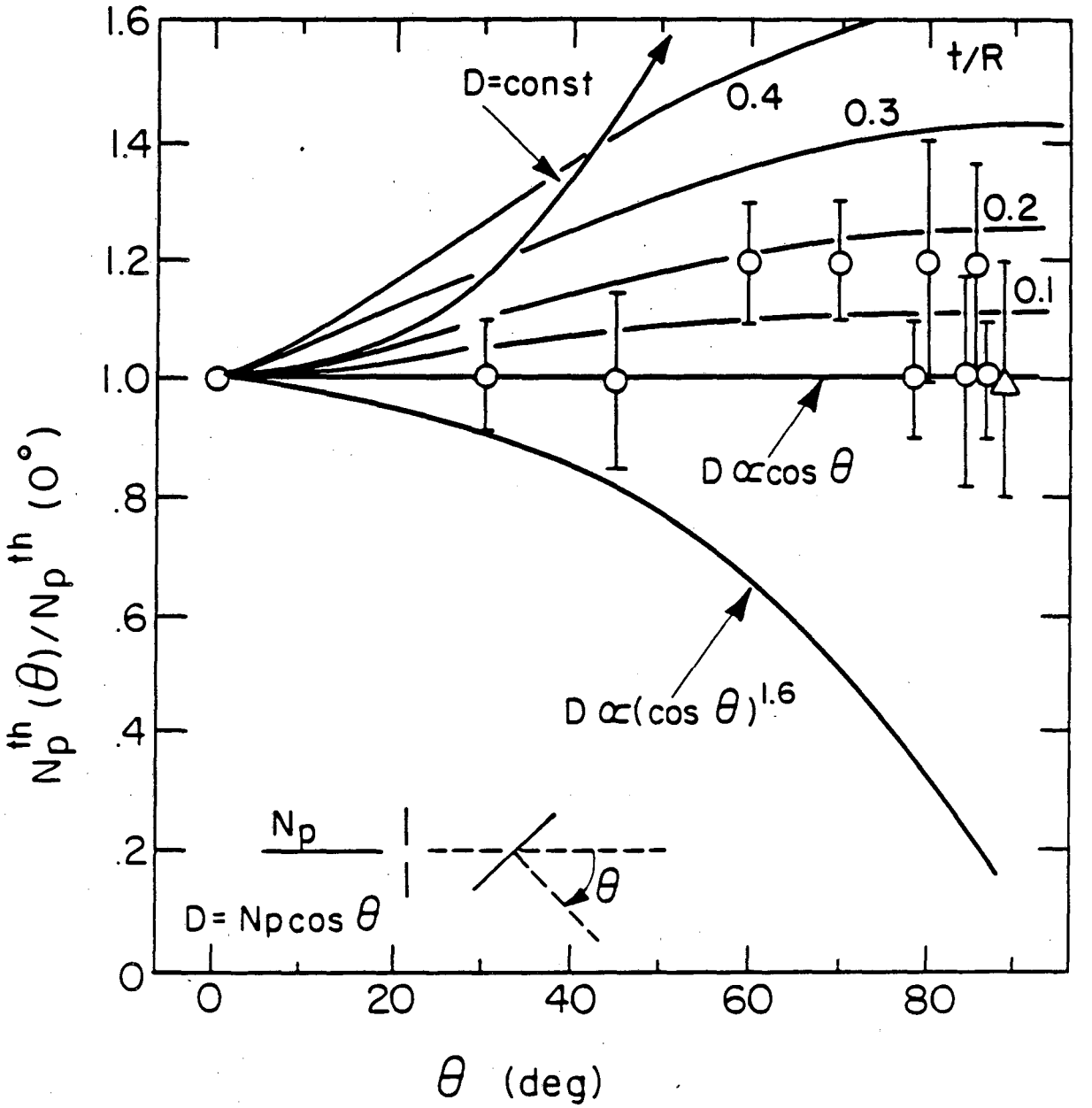

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Fig. 9.

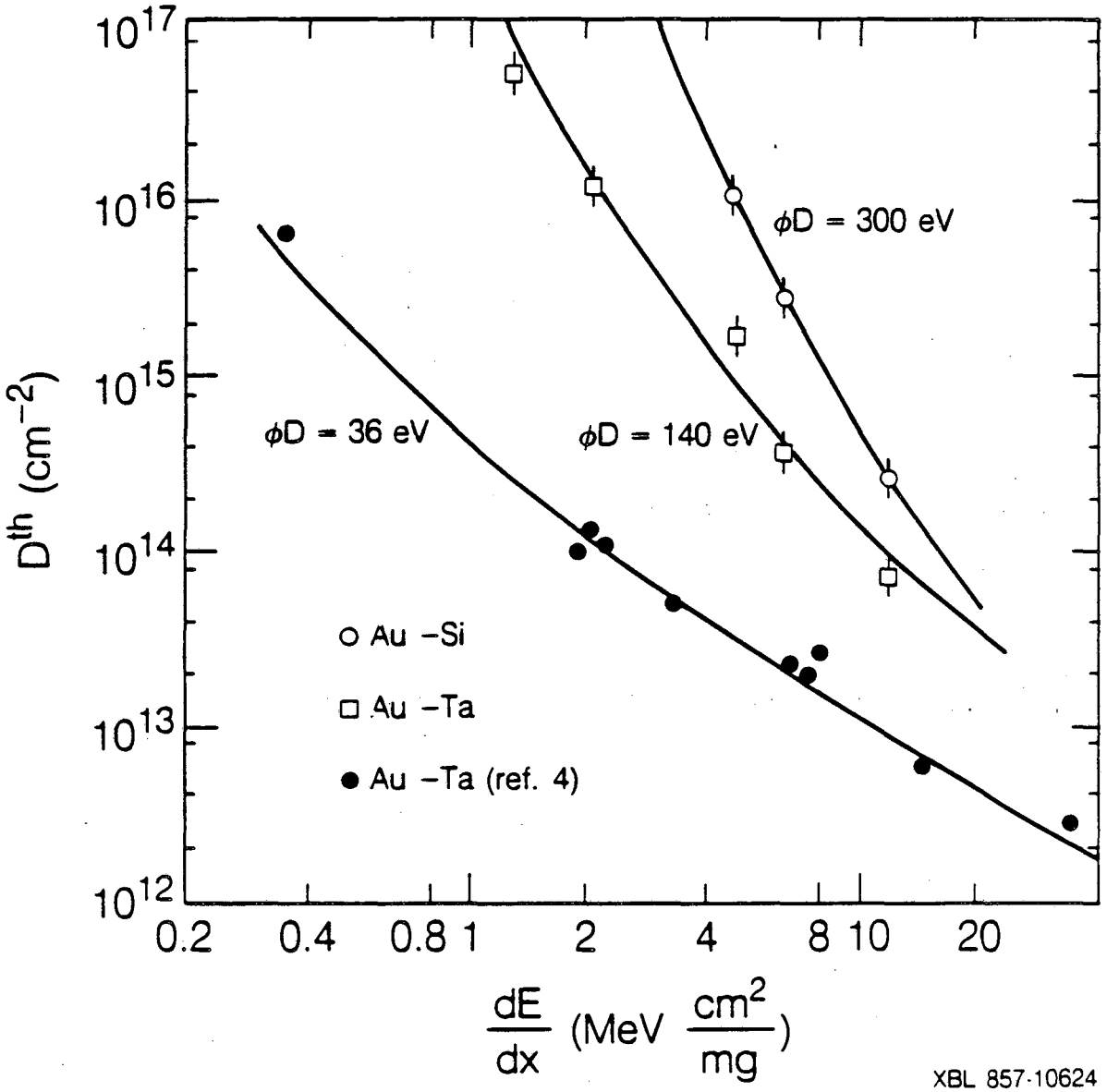
10^{17}
10^{16}
10^{15}
10^{14}
10^{13}
10^{12}
D^{th} (cm^{-2})
ϕD = 300 eV
ϕD = 140 eV
ϕD = 36 eV
○ Au –Si
□ Au –Ta
● Au –Ta (ref. 4)
0.2
0.4
0.8
1
2
4
8
10
20
$\frac{dE}{dx}$ (MeV $\frac{cm^2}{mg}$)
XBL 857-10624

Fig. 10.

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