TRANSMISSION SPUTTERING OF GOLD THIN-FILMS BY
LOW-ENERGY (<1 keV) XENON IONS: I. THE SYSTEM AND THE MEASUREMENT

by

Guy Ayrault and David N. Seidman

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TRANSMISSION SPUTTERING OF GOLD THIN-FILMS BY LOW-ENERGY
(<1 keV) XENON IONS: I. THE SYSTEM AND THE MEASUREMENT[†]

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ABSTRACT

A novel system for direct measurement of the transmission sputtering yields of ion-irradiated thin films is described. The system was specifically designed for the study of the transmission sputtering caused by low-energy (<1 keV) xenon ions. The xenon ion-beam employed was first mass analyzed in a specially-constructed crossed magnetic and electric-field mass spectrometer; this analyzer eliminated all energetic neutral and singly-charged ions of mass less than 40 amu; it is also expected that $\leq 2\%$ of the xenon ions which actually reached a specimen were doubly-charged. The analyzed xenon-ion beam was made to impinge on a gold thin-film (~ 100 to $500 \text{ \AA}$ thick) which was mounted in a JEM 200 transmission electron-microscope holder. The temperature of the specimen could be varied between ~ 25 and 300 K employing a continuous-transfer liquid-helium cryostat. The particles (atoms or ions) ejected from the unirradiated surface of the gold thin-film were detected by two channetron electron-multiplier arrays in the Chevron configuration; the Chevron detector was able to detect individual transmission sputtered particles when operated in the saturated mode. To further enhance resolution the electron cascades, produced by the CEMA, were amplified and shaped electronically into uniform square pulses. The shaped signals were detected with an Ithaco 391A lock-in amplifier (LIA). With the aid of a ratiometer feature in the LIA we were able to measure directly the ratio of the transmission sputtered-current (I_t) to the incident ion-current (I_b); for $I_b = 1 \mu\text{A cm}^{-2}$ a ratio of I_t/I_b as small as 1×10^{-9} has been measured. A detailed discussion of the calibration procedure and the experimental errors, involved in this technique, are also presented.

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1 INTRODUCTION

The concept of focused collision sequences (FCSs) or focusons was introduced to the field of radiation damage by Silsbee⁽¹⁾ in 1957, to explain the role played by a perfect crystal lattice in the transport of momentum. Silsbee demonstrated analytically that FCSs provide an efficient mechanism for the transport of momentum along close-packed crystallographic directions (e.g., the $\langle 110 \rangle$, $\langle 100 \rangle$ and $\langle 111 \rangle$ directions in the face-centered cubic lattice) without the permanent displacement of lattice atoms. Shortly thereafter Vineyard and co-workers⁽²⁾ simulated low-energy (< 1 keV) radiation damage events, in a face-centered cubic copper lattice, employing the molecular-dynamic computer technique. The computer experiments of Vineyard et al. on copper showed the existence of FCSs and also a new physical phenomenon that is now called a replacement collision sequence (RCS); RCSs were observed in both the face-centered and body-centered (α -iron) cubic lattices.⁽²⁻⁶⁾ An RCS involves the transport of mass as well as momentum along close-packed crystal-lattice directions. The RCS mechanism allows for the dynamic transport of a self-interstitial (SIA) a number of interatomic spacings away from its vacant lattice site; the SIA can be transported by this mechanism even at a temperature where it cannot migrate as a result of a thermally-activated mechanism. The Frenkel pair produced in this manner is stable as long as the SIA is located outside of the SIA-vacancy recombination volume.⁽²⁾ Subsequently Seeger⁽⁷⁾ pointed out that the existence of RCSs would result in the transport of SIAs away from the region of a displacement spike^(8,9) to produce a depleted (or diluted) zone. Thus since the early sixties RCSs and FCSs have been discussed extensively as one of the important factors in determining the spatial arrangement of vacant lattice sites and SIAs in irradiated metals [e.g., see Thompson⁽¹⁰⁾, Nelson⁽¹¹⁾, and Seidman⁽¹²⁾]; that is, the initial spatial arrangement before the onset of any thermally-activated point-defect

migration. However, direct experimental measurement of the mean range and range distribution of either FCSs or RCSs in any metal has proved to be an elusive goal. More specifically, different types of experiments on gold[†] have been interpreted as indicating a mean RCS range of $50 \text{ \AA}^{(13)}$, $175 \text{ \AA}$ at $300 \text{ K}^{(14)}$, $1400 \text{ \AA}^{(15)}$, $2000 \text{ \AA}^{(16)}$, and $>4000 \text{ \AA}^{(17)}$. The overall experimental situation for a number of different metals and alloys, with respect to RCSs, has been reviewed by both Venables⁽¹⁸⁾ and Blewitt et al.⁽¹⁹⁾

Sputtering experiments have been used as a technique to study FCSs and RCSs; two general types have been employed. The first type is back sputtering and involves the study of the particles ejected from the irradiated surface of a bulk specimen.^{††} The second type is transmission sputtering and involves the study of the particles ejected from the unirradiated surface as a result of an irradiation with either low-energy heavy-ions [Figure 1(a)] or light energetic-particles [Figure 1(b)]. Transmission sputtering experiments are more direct than back-sputtering studies to analyze, but have not been performed extensively because they are experimentally more difficult. The now classic transmission-sputtering experiment was performed by Nelson and Thompson⁽²³⁾ employing protons that had an incident energy of ~ 2.8 to 3.6 MeV ; the protons that emerged from the back surface of the gold films had a mean energy of $\sim 0.3 \text{ MeV}$. Figure 1(b) illustrates schematically the physical situation for this type of experiment. The incident protons entered the gold thin-film and produced damaged regions (i.e., depleted zones), as a result of elastic collision events, throughout the thickness of the film; for proton damage the depleted zones were very spatially extended.⁽¹²⁾ Both RCSs and FCSs were

[†] We restrict our discussion to gold since this metal has been studied extensively and yet the results have remained controversial for approximately a decade.

^{††} The reader is referred to the books of Thompson,⁽²⁰⁾ Nelson,⁽²¹⁾ and Carter and Colligon⁽²²⁾ for further details and results concerning back-sputtering experiments.

ejected out of each depleted zone and propagated through the thin film. Those that had a sufficient range and a large enough energy to overcome the binding forces at the unirradiated surface produced ejected atoms or ions at this surface of the film. The sputtered material was collected on a substrate and the deposit was analyzed by autoradiography; they observed a well-developed preferential ejection-pattern. From these experiments Nelson and Thompson concluded that the focusing energy, for the $\langle 110 \rangle$ directions in gold, was greater than 350 eV. Recently Bay et al.⁽²⁴⁾ have studied the transmission sputtering, produced by 500 keV Ar^+ ion bombardment, of polycrystalline gold-films as a function of film thickness; Rutherford back-scattering was used to determine the thickness of the film. The sputtering yield versus thickness of the target data fitted the Sigmund⁽²⁵⁾ theoretical curve for transmission sputtering of amorphous materials out to a thickness of 2000 Å. Thus they concluded that long-range RCSs (i.e., greater than 2000 Å) had not contributed to the measured sputtering yields. Kirk and Conner⁽²⁶⁾ have performed an experiment employing fission-neutron sputtering of gold single-crystals to observe the ejection pattern caused by RCSs or FCSs. They claimed that >85% of the transmission-sputtered atoms or ions were ejected randomly; i.e., no evidence of a preferential ejection pattern was observed. Cherns et al.⁽²⁷⁾ have studied the transmission sputtering of (111) gold films (200 to 1000 Å thick) employing 0.4 to 1.15 MeV electrons; in this energy range no permanent radiation damage was produced.[†] The atoms or ions ejected from the unirradiated surface of the films were collected on a substrate and detected by x-ray microanalysis.⁽²⁸⁾ Ejection patterns were observed and they exhibited strong three-fold symmetry. For the very low transferred energies

† For the Cherns et al. experiment no radiation damage was produced; therefore, when thinking about this experiment the damaged regions (depleted zones) must be mentally omitted.

involved in this experiment the ejection patterns must have been caused by FCSs with a range of only one to three interatomic spacings.⁽²⁷⁾

It is emphasized that the four transmission-sputtering experiments discussed above are of the type illustrated in Figure 1(b), where the incident irradiating particles passed deep into or even through the gold thin-film. For the present experimental study of transmission sputtering we have employed the physical arrangement shown in Figure 1(a). In this case the mean-range of the low-energy heavy-ion was always very small compared with the thickness of the gold thin-film, so that all the vacancy type damage was located very close to the irradiated surface. The FCSs and/or RCSs which were ejected from each damaged region (depleted zone) in the forward direction resulted in ejected particles if they had a range greater than the film thickness, and sufficient energy to overcome the binding forces at the unirradiated surface. Figure 2 illustrates the fact that it is, at present, not possible to distinguish between FCSs or RCSs by this transmission sputtering technique. The end product is always an ejected atom or ion and one cannot, at present, determine whether the vacancy was left behind near the irradiated surface (RCS) or at the unirradiated surface (FCS). It should be noted (see Figure 2) that if the range of some or all of the RCSs was less than the thickness of the thin film that SIAs would be deposited within the specimen; furthermore, if the SIAs were mobile at the irradiation temperature they would cluster to form dislocation loops.⁽¹⁴⁾ The only published transmission-sputtering experiment, of the type illustrated in Figure 1(a), had been performed by Ecker.^(13,28) He employed both (100) or (111) gold thin-films and 0.5 keV Ar^+ , 1 keV Kr^+ , or 0.25 to 1 keV Xe^+ ions as the irradiating species; the thicknesses of the gold thin-films ranged from 35 to 150 $\text{\AA}$. For all of these

cases Ecker stated that the measured transmission-sputtering yields were below the detection limit [$(1 \text{ to } 5) \times 10^{-3} \text{ atom ion}^{-1}$] of his technique. From these experiments he concluded that less than 10^{-2} of the FCSs or RCSs produced had traveled more than $50 \text{ \AA}$ along $\langle 110 \rangle$ type directions.

In this two part series on the transmission sputtering of gold thin films, by low-energy (300 or 700 eV) Xe^+ ions, we present a new approach⁽²⁹⁾ to this problem which employs two channeltron electron-multiplier arrays (CEMAs) in the Chevron configuration⁽³⁰⁾ as a particle detector;⁽³¹⁾ for the xenon energies we employed our experiment is of the type illustrated in Figure 1(b). This paper (Part I) presents the apparatus and the measurement technique, while Part II⁽³²⁾ concentrates on experimental results. At the heart of the total system is the Chevron CEMA detector and a lock-in amplifier (LIA) which was used to improve the signal-to-noise ratio; with this combination we have been able to measure transmission-sputtering yields as small as 10^{-9} . The 300 or 700 eV Xe^+ ion beam, employed for the irradiations, was analyzed by a crossed magnetic and electric-field mass-analyzer. The gold thin-film specimens could be irradiated anywhere between $\sim 25 \text{ K}$ and room temperature employing a continuous-transfer liquid-helium cryostat. A transmission electron microscope (TEM) specimen holder was employed to mount each gold film, and after irradiation each film was examined in a TEM to determine its perfection and thickness. A detailed discussion of the calibration procedure and the experimental errors is given in Sections 3 and 4 and Appendix A.2. In addition to being used for transmission sputtering experiments the apparatus described can also be used to study the channeling of specific low-energy light ions (e.g., H^+ or He^+). The detailed results on the transmission sputtering of thin gold-films by 300 or 700 eV Xe^+ ions is presented in Part II.

2 EXPERIMENTAL APPARATUS

2.1 The Detection and Measuring System

Detection of the transmission sputtered particles was accomplished dynamically with a Galileo Chevron detector.⁽³⁰⁾ This (see Figure 3) consisted of two channel electron-multiplier arrays (CEMAs) in a series; the 37.5 μm diameter channels within each CEMA were at an angle of 8° with respect to the surface of the CEMA and at an angle of 16° with respect to the channels in the other CEMA; this arrangement prevented secondary ion feedback.⁽³⁰⁾ The front surface of the first CEMA, in our experimental setup, could be biased at a potential anywhere between -1000 to 900 Vdc with respect to the gold thin film; the potential drop across each CEMA was between 700 to 1000 Vdc. For visual observation of the transmission sputtered particles a phosphor screen,* deposited on a tin-oxide coated circular glass-disc,^(33,34) was placed immediately below the Chevron (see Figure 3). The phosphor screen was maintained at a potential of +2200 Vdc with respect to the bottom surface of the second CEMA when the front surface of the first CEMA was at -1000 Vdc, or at 600 Vdc when this same surface was at 900 Vdc. When the Chevron detector was operated in the saturated mode⁽³¹⁾ an individual transmission sputtered particle could initiate an electron cascade which resulted in a final output of $\sim 10^7$ electrons over a time period of ~ 10 nsec. The amplification of each individual sharp electron-pulse made possible the detection of single transmission sputtering events.

A schematic diagram of the Chevron detector and its relationship to the irradiated gold thin-film is shown in Figure 3. The gold film was mounted in a standard JEM 200 transmission electron microscope specimen holder underneath a 3 mm diameter copper disc with either a 50 or 100 μ diameter hole in its center. An analyzed beam (see Section 2.3) of 300 or 700 eV Xe^+

* A Sylvania No. 160 phosphor (P-1) was employed. This phosphor is $\text{Zn}_2\text{SiO}_4:\text{Mn}$; it has a fluorescent and phosphorescent color in the green and is a medium persistence phosphor.

ions, pulsed at a frequency of 100 Hz, was made to impinge along the normal to the surface of the thin film. In most experiments the gold thin-film was maintained at a potential of +1000 Vdc with respect to the front surface of the first CEMA; hence a positively-charged[†] ejected ion had a parabolic trajectory, while an ejected neutral atom had a straight-line trajectory* (see Figure 3).

The electronic system shown in Figure 4 was employed to measure the transmission sputtering yields. The individual electron-pulses produced by the Chevron were collected from the phosphor screen, then amplified and shaped into uniform square-pulses 0.8 V in amplitude and 10 μ sec wide; the discriminator before the amplifier rejected any small spurious voltage-pulses. The square pulses were then smoothed employing a simple resistance-capacitance (RC) filter with a time constant of 1 msec. Finally the pulses were fed into an Ithaca model 391A lock-in amplifier (LIA). The 300 or 700 eV Xe⁺ ion beam was pulsed at a frequency of 100 Hz and this same value of frequency was used as a reference for the LIA. In this way the signal-to-noise ratio was improved by reducing the influence of random background noise.

The measured transmission sputtering yield (S_{tm}) is defined as

$$S_{tm} = \frac{\text{measured number of Chevron counts}}{\text{measured number of ions striking the thin gold-film.}} \quad (1)$$

A quantity R proportional to S_{tm} was measured directly by a ratiometer which was built into the LIA. The incident ion-current (I) was first fed into a circuit which measured and averaged the ac component of I; then a dc voltage (V_I)

[†] In the case where negatively charged ejected species were detected (see Section 3.4 of Part II) these species also had, of course, a parabolic trajectory.

* This is a first-order qualitative picture of the trajectories of the ejected atoms or ions as the molecular dynamic calculations of Cherns et al. (27) indicated that the exact trajectory of a neutral atom is a complicated function of surface binding energy and the atom's kinetic energy when ejected.

proportional to this ac component was fed to the LIA. In addition the value of V_I , which was nearly constant, was measured digitally and recorded by hand. The ratio (R) of V_{LIA}/I , which was directly proportional to S_{tm} , was measured by the ratiometer.* The values of R and V_{LIA} were recorded as a function of time on a dual-pen strip-chart recorder (see Figure 4). The conversion of the recorded R values into S_{tm} values, and the calibration of the system, is discussed in Section 3.

2.2 Low-Energy Gas-Ion Gun

A schematic drawing of the low-energy gas-ion gun (herein after called an ion gun) is shown in Figure 5. The ion gun consisted of a plasma chamber at earth potential, a resistively-heated tungsten-filament, an extractor lens and a focusing lens: the plasma chamber sat in the center of an axial magnetic field produced by a solenoid.** In practice high-purity (<20 ppm total impurities) xenon gas was bled into the plasma chamber, via a Varian leak valve, and ionized by electrons which were thermionically emitted from the negatively-biased (-45 Vdc) tungsten filament. The axial magnetic field was used to increase the electron path length. The Xe^+ ion beam was extracted and focused employing lenses which were biased at earth and a negative potential respectively. In addition the specimen holder and mass analyzer (see Section 2.3 and Figure 6) were maintained at a negative potential (300 or 700 Vdc); therefore, since the plasma chamber was maintained at earth potential the ion energy at the specimen was determined by this negative potential. To produce a pulsed Xe^+ ion beam the negative bias on the tungsten filament was pulsed to -45 V at 100 Hz with a duty cycle of 0.5 (see Figure 3): this

* The value of V_{LIA} was proportional to the in-phase Chevron count rate averaged over a time period (τ_L) of 12.5 to 125 sec.

**The present ion gun was designed on the basis of several earlier designs reported in the literature.(35-38)

provided a pulsed ion-current which had a rise time of ~ 0.5 msec. At 700 eV the maximum Xe^+ ion-current density at the specimen, approximately 7.5 cm away from the bottom of the focusing lens was $\sim 1 \mu\text{Acm}^{-2}$, while for the 300 eV Xe^+ ions the maximum current density was $\sim 0.4 \mu\text{Acm}^{-2}$.

2.3 The Mass Spectrometer

Initially we performed several irradiations with an earlier version of this system which employed an unanalyzed ion beam and also subsequently with a magnetically analyzed beam from which light ions were removed, but not energetic neutral atoms. For both arrangements we observed values of S_{tm} which were $> 10^{-8}$ for films as thick as $800 \text{ \AA}$. These signals were most likely due to the channeling of light ion and energetic neutral⁽³⁹⁾ impurity-atoms (e.g., H, H^+ , He and He^+); with the present mass analyzer (crossed electric and magnetic fields) these signals disappeared.

To eliminate completely the possibility of light ions and/or energetic neutrals from reaching the gold thin-film the Xe^+ ion beam was analyzed with the mass spectrometer shown in Figure 6. The ion beam passed through the first and second apertures, 0.254 cm in diameter, before it entered the analyzing section of the mass spectrometer. Next, the ion beam passed through a pair of parallel electrostatic deflection plates at a potential $V_0 \pm \frac{V'}{2}$, where V_0 was the potential of the ion beam and V' was the potential difference across the plates. Finally, the ion beam passed through a magnetic deflector which consisted of a permanent magnet with soft-iron pole pieces*: a field of ~ 2.5 kgauss was required to deflect a 700 eV Xe^+ ion beam through 8° .[†] When the mass spectrometer was adjusted for an Xe^+ ion beam

* The magnetic field-vector is at a right angle with respect to the electric field-vector.

† The total deflection angle in going through the electrostatic and magnetic analyzing sections was 17° .

our design prevented all energetic neutrals and singly-charged ions of mass less than 40 amu from reaching the specimen. The analyzed Xe^+ beam then passed through a third 0.254 cm diameter defining aperture before impinging on the gold thin-film. The trajectories of an H^+ impurity ion-beam and an energetic neutral atomic beam are also indicated in Figure 6.

In our experiments the Xe atoms were ionized with 45 eV electrons; at this energy it was anticipated that ~5% of Xe^+ ions should have been doubly-charged.⁽⁴⁰⁾ However, an analysis of our mass analyzer indicated that with the electrostatic deflector properly adjusted for Xe^+ ions, that at most 40% of the doubly-charged species were able to reach the specimen. Therefore, we expected that <2% of the xenon ions which actually reached the specimen were doubly-charged.

The energy spread of the ion beam was measured for both the 300 and 700 eV Xe^+ ions. The technique was similar to that used for calibration of the ion current density (see Section 3); i.e., the thin film specimen was replaced by a small aperture, and the ion current through this aperture was collected and measured, with a variable retarding potential applied to the collector. It was found for both the 300 and 700 eV Xe^+ ions that the total energy spread was ≤ 8 eV. By this we mean that the total change in the retarding potential between the point where the measured current began to decrease, and the point where the measured current went to zero, was ≤ 8 Vdc.

2.4 The Total Sputtering System

Figure 7 shows the entire sputtering system in its vacuum system. All the components of the sputtering system were suspended from a circular stainless steel plate, for easy demounting, which rested on a glass cylinder. The latter in turn rested on a second circular stainless-steel plate. The glass cylinder sat inside a fine steel-mesh cylinder which served as a Faraday cage. The vacuum system was first rough pumped with a vacsorb pump, to prevent oil contamination, and then pumped with an oil diffusion pump, 5.08 cm diameter, which was trapped with a Granville-Phillips liquid-nitrogen

cold-trap. The background pressure achieved routinely with this system was $\sim 10^{-7}$ Torr.*

The copper specimen-mounting block was cooled by conduction using a continuous-transfer liquid-helium cryostat⁽⁴¹⁾ with which the specimen was maintained at any temperature between ~ 25 and 300 K. The entire cryostat was electrically isolated from the copper specimen mounting-block by a piece of single crystal Al_2O_3 (sapphire). The specimen temperature was measured with a platinum resistance thermometer (item H in Figure 7) which was attached to the mounting block. The gold thin-film was tightly clamped to the copper specimen-holder with the aid of a JEM 200 specimen holder (see Figure 3). This arrangement guaranteed that the gold-thin-film strip was at the temperature measured by the platinum resistance thermometer. In Appendix A.1 we show that the maximum possible temperature rise in the thin film due to ion-beam heating was $\sim 3 \times 10^{-3}$ K for the geometry employed. The front-surface mirror shown in Figure 7 was used to observe the pattern produced by the transmission-sputtered particles.

3. CALIBRATION AND DATA ANALYSIS

The physical quantity of interest in the transmission-sputtering experiments is S_{tm} as a function of the thickness of a gold thin-film. To determine S_{tm} from the measured quantities V_{LIA} and I we had to know λ_s as a function of V_{LIA} , the local current density (j) striking the thin film as a function of I , and the irradiated area (A) of the thin film. The quantity V_{LIA} as a function of λ_s was determined by replacing the sharp electron pulses from the Chevron detector with a pulse generator which operated at a known frequency and was gated by a square-wave generator operated at the 100 Hz ion beam frequency. The LIA output V_{LIA} as a function of λ_s was found to be linear over the normal operating range for these experiments (~ 1 to 1000 Hz);

* The background pressure was mainly xenon since the liquid-nitrogen cold-trap cryopumped this gas.

i.e.,

$$V_{LIA} = \kappa \lambda_s \quad (2)$$

where λ_s was the measured count rate (number count sec^{-1}) and κ was a calibration constant.

The quantity j as a function of I was determined by replacing the gold thin film by a disc with a precisely measured aperture and simultaneously measuring both I and the ion current (I') through the aperture. Hence the value of j was given by

$$j = I'/A_a, \quad (3)$$

where A_a was the area of the aperture; the quantity j was also given by

$$j = \xi I; \quad (4)$$

where

$$\xi = I'/(IA_a) \quad (5)$$

is a second calibration constant. The value of A was determined by measuring the diameter of the hole in the copper mounting disc using an optical microscope with a micrometer eyepiece; the microscope and eyepiece were calibrated using a standard glass-grating. The quantity S_{tm} defined by Eq. (1) in terms of practical quantities was given by

$$S_{tm} = 1.60 \times 10^{-19} \lambda_s / (jA) \quad (6)$$

and using Eqs. (2) and (4), Eq. (6) became

$$S_{tm} = 1.60 \times 10^{-19} (V_{LIA}/I) / (\kappa \xi A), \quad (7)$$

where the numerical factor converts I in amperes to number of ions per sec.

From the above it is seen that S_{tm} could be determined as a function of time (t) by reading the value of the quantity $R = (V_{LIA}/I)$ off the strip-chart recorder and then multiplying it by the constant $1.60 \times 10^{-19} / (\kappa \xi A)$. The quantity S_{tm} increased with t as the thin film was back-sputter thinned by the Xe^+ ion-beam; the back-sputtering rate was enormous compared to the transmission sputtering rate. To determine S_{tm} as a function of the film

thickness, we had to know the total ion-dose (Φ) along with any two of the following three quantities: (1) the initial thickness of thin film; (2) the final thickness of the thin film; and (3) the sputtering rate.

For simplicity we assumed that the sputtering rate was not a function of Φ , although this is not necessarily the case.⁽⁴²⁾ Even with this assumption the determination of any of the above quantities presented certain difficulties. As described in the following paper the thickness of each thin film was determined by measuring, by transmission electron-microscopy, the projected widths of stacking faults which lie on the (111) planes. Because of the inevitable carbon contamination of the thin films, in the electron microscope,⁽⁴³⁾ it was found to be undesirable to measure the thickness of the thin film prior to its irradiation in the sputtering system. If the as-deposited films were perfectly uniform, measurement of a nearby section of the same as-deposited film would be adequate. Unfortunately, some of the as-deposited films exhibited fairly large long-range variations in thickness. Moreover transmission sputtering yields from different specimens cut from the same as-deposited films were not in good agreement if one assumed an identical initial thickness. The agreement was found to be better if we measured the final film-thickness by TEM and used our own measured value of ~ 2.0 atom ion⁻¹ at 700 eV for the backsputtering rate (the most commonly used value of energy in our experiments). At 300 eV our own data was inadequate to determine a back-sputtering rate, so we assumed that the scaling with bombardment energy was the same as for the published data*; this procedure yielded a value of ~ 1 atom ion⁻¹ at 300 eV.

* Back-sputtering yield data are available for Xe⁺ on polycrystalline Au for bombardment energies up to 600 eV.⁽⁴⁴⁾ An extrapolation of this data to 700 eV yielded a value of ~ 3.7 atom ion⁻¹. This value was obtained for very high Xe⁺ doses on Au and for polycrystalline specimens. Thus we attributed the difference between our value of the backsputtering rate and the Rosenberg and Wehner (44) value to differences in the experimental conditions.

4 EXPERIMENTAL ERRORS

The two major sources of measurement error in our experiments were fluctuations in j and statistical scatter. The former error was peculiar to our experimental set-up, while the latter one was intrinsic to the experiment. In Section 3 we discussed the relationship between j and I and showed that if j was uniform then the calibration constant ξ should have been independent of operating conditions; unfortunately it was not uniform. Small changes in the ion-gun operating conditions changed ξ by as much as 30%, although 10% changes were more typical. The changes in ξ caused similar changes in the output ratio R .

In Appendix A.2 we present the derivation of an equation for the variance of V_{LIA} ($\text{var}[V_{LIA}] = \sigma_V^2$) under the assumption that V_{LIA} and the noise were both distributed according to a Poisson distribution. We showed (see Appendix A.2) that the ratio of $(\sigma_V/E_s\{V_L\})$ is given by

$$\frac{\sigma_V}{E_s\{V_{LIA}\}} \approx \frac{1}{\sqrt{RC}} \left[\frac{1}{\lambda_s} + \frac{2\lambda_n}{\lambda_s^2} \right]^{1/2}, \quad (\text{A.2.29})$$

where $E_s\{V_{LIA}\}$ is the expectation value of V_{LIA} , R and C are the resistance and capacitance values of the RC filter* in the LIA output section, and λ_s and λ_n are the mean pulse-frequencies of the signal and noise respectively. As expected σ_V is inversely proportional to the square-root of RC (i.e., τ_L). Also if λ_s is $\gg \lambda_n$ then the noise term can be neglected, but if λ_s is $\ll \lambda_n$ then the statistical scatter term dominates σ_V .

The quantity τ_L was a variable, therefore we could have, in principle, reduced σ_V to any arbitrary extent by increasing τ_L (i.e., the quantity RC). This approach had several drawbacks. First, since we wished to measure an output as a function of film thickness as the film was sputter thinned, or equivalently as a function of t , the quantity τ_L had to be maintained at a value much less than any

* Note that quantity RC is the characteristic averaging time (τ_L).

characteristic time involved in the change in the output. Second, a large value of τ_L implied a long transient response time when the Xe^+ ion beam was turned on. Because of problems in experiments we had performed earlier, connected with synchronous background signals unrelated to transmission sputtering, the Xe^+ ion beam was regularly deflected away from the specimen to check for a possible recurrence of this effect. To avoid the necessity of discarding a lot of data when the irradiation was re-initiated a good transient response (i.e., small τ_L) was found to be essential. A third problem with a large value of τ_L involved an experimental error which we have not yet discussed. Although a good deal of care was taken to shield the detection system from external noise the shielding was not perfect and bursts of noise, from nearby laboratories, occasionally intruded. The presence of these noise bursts was easily detected with an auxiliary auditory system and was noted on the strip chart recording of the output. The noise bursts were typically more intense than the real output and they were capable of causing considerable deviations in V_{LIA} . For small values of τ_L it was practical to ignore the data for a few τ_L after the noise burst. As τ_L was increased more and more data could have been lost in this way. In most experiments a τ_L value of either 25 or 80 sec was used, although in a few experiments a τ_L of 250 sec proved necessary; the largest value of τ_L did cause difficulties which will be discussed later in Part II. (32)

5 SUMMARY

A novel transmission sputtering system was described. It had the following features:

1. The particles (atoms or ions) ejected from the unirradiated surface of an irradiated thin-film, as a result of transmission sputtering, were detected dynamically with a Galileo Chevron detector. When the Chevron detector was operated in the saturated mode individual transmission-sputtered particles produced sharp electron-pulses. In addition the transmission-sputtering

pattern produced was observed with the aid of a phosphor screen placed directly behind the Chevron detector.

2. The irradiating ion-beam (<1 keV Xe^+ ions) was pulsed at a known frequency (typically 100 Hz) and this same value of frequency was used as a reference for an Ithaco model 391A lock-in amplifier (LIA). In this way the signal-to-noise ratio was improved by reducing the influence of the random background noise.

3. The LIA had a ratiometer feature which allowed the direct measurement of the ratio of the transmission sputtered current (I_t) to the incident ion-current (I_b); for $I_b = 1 \mu\text{A cm}^{-2}$ a ratio of I_t/I_b as small as 10^{-9} has been measured.

4. The Xe^+ ion-beam used for the sputtering experiments was mass analyzed in a crossed electronic and magnetic field analyzer. With the mass spectrometer adjusted for an Xe^+ ion beam our design prevented all energetic neutrals and singly-charged ions of mass less than 40 amu from reaching the gold thin-film. An analysis of the mass spectrometer indicated that $\leq 2\%$ of the xenon ions which actually reached the specimens were doubly-charged.

5. The gold thin-film specimen was mounted on a JEM 200 transmission electron microscope holder. The temperature of the specimen could be maintained at any value between ~ 25 and 300 K by means of a continuous-transfer liquid-helium cryostat. After each irradiation the specimen was transferred to a Siemens 102 TEM where its thickness was determined and its perfection was characterized.

6. The calibration and data analysis procedures were given and a detailed analysis of the experimental errors involved was presented.

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APPENDICES

A. 1 Heating of the Specimen During the Ion Irradiation

The irradiated portion of the gold thin-film had the geometry of a small right-circular cylinder with a radius r_0 that was either 25 or 50 μ and a height (ℓ_0) that had a range from 10^{-6} to 4×10^{-6} cm. This cylindrically-shaped volume was situated in the center of a strip of gold thin-film that was ~ 0.1 and ~ 0.3 cm on each side. The surrounding strip of gold thin-film was so well clamped in the specimen holder that we assumed that its temperature was unaffected by the irradiation (see Figure 3). The governing steady-state heat equation for this problem is

$$\frac{1}{r} \frac{d}{dr} \left[r \frac{dT}{dr} \right] + \frac{Q}{\ell_0 K} = 0, \quad (\text{A.1.1})$$

where r is the radial direction in the plane of the thin-film, T is the temperature, Q is the uniform and constant surface heating density (w cm^{-2}) and K is the thermal conductivity. The ratio (r_0/ℓ_0) was always much greater than unity so we treated the heat-flow problem as a two-dimensional one in the plane of the thin film. The boundary conditions used were: (1) the temperature was a constant at $r = r_0$ and equal to the temperature measured by the platinum resistance thermometer; and (2) the slope (dT/dr) at $r = 0$ was equal to zero. The integration of Eq. (A. 1. 1) twice and the evaluation of the constants of integration employing the above boundary conditions yielded the following expression for the maximum temperature rise:

$$\Delta T_{\max} = r_0^2 Q / (4 \ell_0 K). \quad (\text{A.1.2})$$

Employing $K = 1.5 \text{ w cm}^{-1} \text{K}^{-1}$, $\ell_0 = 10^{-6} \text{ cm}$, $r_0 = 5 \times 10^{-3} \text{ cm}$, and $Q = 7 \times 10^{-4} \text{ w cm}^{-2}$ we calculated a $\Delta T_{\max} \approx 3 \times 10^{-3} \text{ K}$. The value of Q was calculated for a 700 Vdc Xe^+ ion beam with a current density of $1 \times 10^{-6} \text{ amp cm}^{-2}$ at the specimen. The physical arrangement of the specimen relative to the ion gun (see Figure 6) was such that the thin film was completely shielded from any radiant heating that may have been caused by the Joule heating of the tungsten filament.

A.2 An Error Analysis of the Statistical Scatter in the Measured Transmission Sputtering Yield (S_{tm})

To calculate the expected statistical scatter in the measured values of S_{tm} (see Section 3) we employed a simple model for the LIA. As shown in Figure 8 we took the LIA to be simply a voltage inverter with a switch and an RC filter; the switch was controlled by the ion-beam pulser. When the Xe^+ ion-beam was on the signal passed directly to the RC filter. Alternatively when the Xe^+ ion-beam was off the signal was inverted. For the purpose of the calculation this configuration was employed to represent an ideal LIA.

The Xe^+ ion-beam was pulsed as shown in Figure 9a. Each ion-beam pulse initiated a number of randomly distributed signal pulses as indicated schematically in Figure 9b. In addition to the signal pulses there was a random distribution of noise pulses as shown in Figure 9c. The sum of the signal plus the noise pulses, through the inverter and switch, gave the voltage pulses V_f exhibited in Figure 9d.

The signal and noise pulses that reached the LIA were all identical and their time duration was short compared to the RC time constant (τ_L) of the LIA. Therefore we ignored the actual shape of the signal and noise pulses and replaced them by delta functions; that is, a single pulse $P(t_0)$ at time t_0 was represented by

$$P(t_0) = P_0 \delta(t - t_0), \quad (A.2.1)$$

where P_0 is the strength of the pulse; P_0 is a constant because of the pulse shape. The voltage at the RC filter due to the signal (V_{fs}), was represented by a steady stream of random pulses $S(t)$ modulated by a function $g(t)$; therefore V_{fs} was represented by

$$V_{fs} = g(t)S(t) \quad (A.2.2)$$

where

$$S(t) = \sum_k P_o \delta(t - T_k) \quad (A.2.3)$$

and

$$g(t) = \begin{cases} 1 & nT < t < (n + \frac{1}{2}) T \\ 0 & (n + \frac{1}{2})T < t < (n + 1) T \end{cases} \quad (A.2.4a)$$

$$(A.2.4b)$$

where $g(t)$ represents mathematically the fact that the Xe^+ ion-beam was pulsed (see Figure 9a) with a period of $(T/2)$ and the T_k are the random arrival times of the signal pulses; the quantity n is an integer ranging from 0 to ∞ . Similarly the voltage at the RC filter due to noise (V_{fN}) was represented by

$$V_{fN} = m(t)N(t) \quad (A.2.5)$$

where

$$N(t) = \sum_j P_o \delta(t - T_j) \quad (A.2.6)$$

and

$$m(t) = \begin{cases} 1 & nT < t < (n + \frac{1}{2}) T \\ -1 & (n + \frac{1}{2})T < t < (n + 1) T \end{cases} \quad (A.2.7a)$$

$$(A.2.7b)$$

where $m(t)$ represents the inverter and switch; compare Eqns. (A.2.4) and (A.2.7).

The total voltage V_f was then given by

$$V_f = V_{fs} + V_{fN} \quad (A.2.8)$$

From the above information we were then able to write the output voltage ($V_{LIA}(t)$) as

$$V_{LIA}(t) = \int_{-\infty}^t e^{-\alpha(t-\tau)} V_{LIA}(\tau) d\tau$$

$$= \int_{-\infty}^t e^{-\alpha(t-\tau)} [S(\tau)g(\tau) + N(\tau)m(\tau)] d\tau; \quad (A.2.9)$$

where α is equal to $1/RC$ and the $\exp[-\alpha(t-\tau)]$ term represents the modulation of the pulses by the large RC of the circuit.

The random arrival times T_k and T_j of the signal and noise respectively, were assumed to be governed by Poisson distributions. Therefore for steady-state conditions the expectation values $E\{S(t)\}$ and $E\{N(t)\}$ of $S(t)$ and $N(t)$ are known to be given by: ⁽⁴⁵⁾

$$E\{S(t)\} = P_o \lambda_s \quad (\text{A.2.10})$$

$$E\{N(t)\} = P_o \lambda_n \quad (\text{A.2.11})$$

where λ_s and λ_n are the mean pulse and signal frequencies for the signal and noise respectively. The expectation value of $V_{LIA}(t)$ was then given by

$$\begin{aligned} E\{V_{LIA}(t)\} &= \int_{-\infty}^t e^{-\alpha(t-\tau)} [g(\tau)E\{S(\tau)\} + m(\tau)E\{N(\tau)\}] d\tau \\ &= \int_{-\infty}^t e^{-\alpha(t-\tau)} [P_o \lambda_s g(\tau) + P_o \lambda_n m(\tau)] d\tau \\ &= E_s\{V_{LIA}(t)\} + E_n\{V_{LIA}(t)\} \end{aligned} \quad (\text{A.2.12})$$

where

$$E_s\{V_{LIA}(t)\} = P_o \lambda_s \int_{-\infty}^t e^{-\alpha(t-\tau)} g(\tau) d\tau, \quad (\text{A.2.13a})$$

and

$$E_n\{V_{LIA}(t)\} = P_o \lambda_n \int_{-\infty}^t e^{-\alpha(t-\tau)} d\tau. \quad (\text{A.2.13b})$$

The integrals in Eqns. (A.2.13) may be evaluated in a similar fashion. First consider Eq. (A.2.13a) for the interval $nT < t < (n + \frac{1}{2})T$; it is given by

$$\begin{aligned} E_s\{V_{LIA}(t)\} &= P_o \lambda_s e^{-\alpha t} \left[\int_{nT}^t e^{\alpha \tau} d\tau + \frac{(n-\frac{1}{2})T}{(n-1)T} \int_{\frac{(n-1)T}{2}}^{(n-\frac{1}{2})T} e^{\alpha \tau} d\tau + \frac{(n-\frac{3}{2})T}{(n-2)T} \int_{\frac{(n-2)T}{2}}^{(n-\frac{3}{2})T} e^{\alpha \tau} d\tau + \dots \right] \\ &= P_o \lambda_s e^{-\alpha t} \left[\int_{nT}^t e^{\alpha \tau} d\tau + \sum_{k=1}^{\infty} \frac{(n-k)T + T/2}{(n-k)T} \int_{\frac{(n-k)T}{2}}^{(n-k+\frac{1}{2})T} e^{\alpha \tau} d\tau \right]. \end{aligned} \quad (\text{A.2.14})$$

Equation (A.2.14) was constructed with the aid of Fig. 9a by considering the time t between nT and $Nt + T/2$. Equation (A.2.14) was evaluated with the aid

of the sum

$$\sum_{k=1}^{\infty} (e^{-\alpha T})^k = e^{-\alpha T} / (1 - e^{-\alpha T})$$

to yield

$$E_s \{V_{LIA}(t)\} = \frac{P_o \lambda_s}{\alpha} [1 - e^{\alpha(nT-t)} \left(\frac{1 - e^{-\alpha T/2}}{1 - e^{-\alpha T}} \right)] . \quad (A.2.15)$$

In a similar manner we obtained

$$E_n \{V_{LIA}(t)\} = \frac{P_o \lambda_n}{\alpha} [1 - 2e^{\alpha(nT-t)} \left(\frac{1 - e^{-\alpha T/2}}{1 - e^{-\alpha T}} \right)] \quad (A.2.16)$$

where the factor of two difference is due to Eqns. (A.2.7). Since the quantity αT was $\ll 1$ Eqns. (A.2.15) and (A.2.16) reduced to

$$E_s \{V_{LIA}(t)\} \approx P_o \lambda_s / (2\alpha) \quad (A.2.17)$$

and

$$E_n \{V_{LIA}(t)\} \approx P_o \lambda_n (t - nT). \quad (A.2.18)$$

It was clear, that since $1/(2\alpha)$ was large compared to $(t - nT)$, that $E_n \{V_{LIA}(t)\}$ was $\ll E_s \{V_{LIA}(t)\}$. Employing Eqn. (A.2.17) we experimentally determined P_o by measuring V_{LIA} for a known λ_s .

The variance of V_{LIA} , $\text{var} [V_{LIA}] = \sigma_V^2$, is given by the usual relationship

$$\text{var} [V_{LIA}] = E\{V_{LIA}^2\} - [E\{V_{LIA}\}]^2 ; \quad (A.2.19)$$

the quantities $V_{LIA}^2(t)$ and $E\{V_{LIA}^2\}$ are given by

$$V_{LIA}^2(t) = \int_{-\infty}^t d\tau \int_{-\infty}^t dv e^{-\alpha(t-\tau)} e^{-\alpha(t-v)} [g(\tau)S(\tau) + m(\tau)N(\tau)] \cdot [g(v)S(v) + m(v)N(v)] \quad (A.2.20)$$

and

$$\begin{aligned}
 E\{V_{LIA}^2(t)\} = & \int_{-\infty}^t d\tau \int_{-\infty}^t dv e^{-\alpha(t-\tau)} e^{-\alpha(t-v)} [g(\tau)g(v) E\{S(\tau)S(v)\} \\
 & + m(\tau)m(v) E\{N(\tau)N(v)\} \\
 & + 2g(\tau)m(v) E\{S(\tau)N(v)\}], \quad (A.2.21)
 \end{aligned}$$

where τ and v are dummy integration variables. The functions $S(\tau)$ and $N(v)$ are statistically independent, therefore

$$E\{S(\tau)N(v)\} = E\{S(\tau)\}E\{N(v)\} = P_O^2 \lambda_s \lambda_n \dots \quad (A.2.22)$$

Also it is known that⁽⁴⁵⁾ the two auto-correlations $E\{S(\tau)S(v)\}$ and $E\{N(\tau)N(v)\}$ are given by

$$E\{S(\tau)S(v)\} = P_O^2 \lambda_s^2 + \lambda_s P_O^2 \delta(\tau - v) \quad (A.2.22a)$$

and

$$E\{N(\tau)N(v)\} = P_O^2 \lambda_n^2 + \lambda_n P_O^2 \delta(\tau - v). \quad (A.2.22b)$$

Therefore, Eqn. (A.2.21) became

$$\begin{aligned}
 E\{V_{LIA}^2(t)\} = & \int_{-\infty}^t d\tau \int_{-\infty}^t dv e^{-\alpha(t-\tau)} e^{-\alpha(t-v)} [P_O^2 \lambda_s^2 g(\tau)g(v) \\
 & + \lambda_s P_O^2 g(\tau)g(v)\delta(\tau - v) + P_O^2 \lambda_n^2 m(\tau)m(v) \\
 & + \lambda_n P_O^2 m(\tau)m(v)\delta(\tau - v) + 2 g(\tau)m(v)P_O^2 \lambda_s \lambda_n]. \quad (A.2.23)
 \end{aligned}$$

Comparing Eqn. (A.2.23) with Eqn. (A.2.12) it was clear that

$$\begin{aligned}
 E\{V_{LIA}^2(t)\} = & [E\{V_{LIA}(t)\}]^2 + \int_{-\infty}^t d\tau \int_{-\infty}^t dv e^{-\alpha(t-\tau)} e^{-\alpha(t-v)} \\
 & \cdot [\lambda_s P_O^2 g(\tau)g(v)\delta(\tau - v) + \lambda_n P_O^2 m(\tau)m(v)\delta(\tau - v)], \quad (A.2.24)
 \end{aligned}$$

and finally from (A.2.19) and integrating over v we obtained

$$\text{var } [V_{LIA}(t)] = \int_{-\infty}^t d\tau e^{-2\alpha(t-\tau)} [\lambda_s P_O^2 [g(\tau)]^2 + \lambda_n P_O^2 [m(\tau)]^2]; \quad (A.2.25)$$

employing

$$[g(\tau)]^2 = g(\tau) \quad (A.2.26a)$$

and

$$[m(\tau)]^2 = 1; \quad (\text{A.2.26b})$$

see Eqns. (A.2.4) and (A.2.7) to understand Eq. (A.2.26). The integration of Eqn. (A.2.25) employing Eqns. (A.2.15) and (A.2.26) yielded

$$\text{var } [V_{LIA}(t)] = \frac{\lambda_s P_o^2}{2\alpha} [1 - e^{2\alpha(nT-t)} \left(\frac{1 - e^{-\alpha T}}{1 - e^{-2\alpha T}} \right)] + \frac{\lambda_n P_o^2}{2\alpha}. \quad (\text{A.2.27})$$

Since $\alpha T \ll 1$ Eqn. (A.2.27) reduced to

$$\text{var } [V_{LIA}(t)] = \sigma_V^2 = \frac{\lambda_s P_o^2}{4\alpha} + \frac{\lambda_n P_o^2}{2\alpha}. \quad (\text{A.2.28})$$

Finally employing Eqn. (A.2.17) we obtained for the expected relative deviation in V_{LIA}

$$\frac{\sigma_V}{E_s \{V_{LIA}\}} \cong \alpha^{1/2} \left[\frac{1}{\lambda_s} + 2 \frac{\lambda_n}{\lambda_s} \right]^{1/2} \cong \frac{1}{\sqrt{RC}} \left[\frac{1}{\lambda_s} + 2 \frac{\lambda_n}{\lambda_s^2} \right]^{1/2}. \quad (\text{A.2.29})$$

FIGURE CAPTIONS

- Figure 1 (a) A schematic diagram illustrating the general principle of transmission sputtering employing low-energy heavy-ions. Note that in this situation the range of the incident ions is much less than the thickness of the evaporated film. See Fig. 2 for a more detailed picture of how particles are ejected from the unirradiated surface of the film as a result FCSs or RCSs.
- (b) A schematic diagram illustrating the general principle of transmission sputtering when a light energetic-particle is employed. In this case the incident ions penetrate deeply into the thin film, producing damaged regions throughout the thickness of the film, the incident particles can also pass through the thin film.
- Figure 2 A schematic diagram illustrating some of the important physical processes that occur when a thin film is irradiated with 300 or 700 eV Xe^+ ions. The film is only 16.32 Å thick ($4a_0$) in this figure, whereas in practice it was 100 to 400 Å thick. The incident Xe^+ ion beam could not traverse the thickness of the film and the Xe ions came to rest within the first few layers of the irradiated surface. The maximum energy transferred to a gold lattice atom was 0.96 of the incident ion energy. Within the first few layers both RCSs and FCSs were produced; for illustrative purposes we have only indicated the ones which were produced along the $[0\bar{1}1]$ and $[011]$ directions. If the range of the FCSs or RCSs was greater than the thickness of film an atom or ion would have been ejected at the unirradiated surface. Note that each RCS left behind a vacancy at the irradiated surface, while a FCS left behind a vacancy at the unirradiated surface.

- Figure 3 A detailed diagram showing the relationship between the thin gold-film and the Chevron detector. An image of the electron signal produced by the Chevron was observed with the aid of the P-1 phosphor screen that was deposited on the tin-oxide coated glass plate.
- Figure 4 A schematic diagram of the electronic circuit employed to measure the transmission sputtering-yields. This circuit is described in detail in Section 2.1.
- Figure 5 The low-energy gas-ion gun used to produce either 300 or 700 eV Xe^+ ions. The extracted and focused Xe^+ ion beam was first made to pass through two beam defining-apertures before it was allowed to enter the mass-spectrometer section of the apparatus (see Figure 6).
- Figure 6 A schematic diagram of the mass spectrometer and its relationship to the ion gun and thin-film specimen. Note that the electric and magnetic field vectors are at right angles to one another. The trajectories of an energetic neutral, an Xe^+ ion, and a H^+ ion are exhibited to illustrate the filtering capability of the mass spectrometer.
- Figure 7 The entire sputtering system shown in its vacuum system. The labeled components are as follows: (A) focusing lens; (B) first, second and third apertures; (C) electrostatic shield; (D) electrostatic deflection plates; (E) permanent magnet; (F) single-crystal Al_2O_3 (sapphire); (G) copper conduction-arm; (H) platinum resistance thermometer; (I) JEM 200 specimen holder; (J) channeltron electron-

multiplier arrays (CEMAs); (K) phosphor screen on a tin-oxide coated glass window; and (L) liquid-helium cryostat.

Figure 8 The schematic model of the lock-in amplifier employed to calculate the expected statistical scatter in the transmission sputtering yield.

Figure 9 (a) A schematic wave-form of the Xe^+ ion-beam current (I) as a function of time (t). The 300 or 700 eV Xe^+ ion beam was pulsed at a frequency of 100 Hz:

- (b) The randomly distributed voltage signal pulses as a function of t ;
- (c) The randomly distributed voltage noise pulses as a function of t ;
- (d) The sum of the signal plus the noise pulses, through the inverter and switch, as a function of t .

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9