Transmission sputtering of gold thin films by low-energy (< 1 keV) xenon ions. I. The system and the measurement

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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 is first mass-analyzed in a specially constructed crossed magnetic- and electric-field mass spectrometer; this analyzer eliminates all energetic neutral and singly charged ions of mass less than 40 amu; it is also expected that <2% of the xenon ions which actually reach a specimen are doubly charged. The analyzed xenon ion beam is made to impinge on a gold thin film (~ 100–500 Å thick) which is mounted in a JEM 200 transmission electron-microscope holder. The temperature of the specimen can 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 are detected by two channeltron electron-multiplier arrays (CEMA) in the Chevron configuration; the Chevron detector is able to detect individual transmission-sputtered particles when operated in the saturated mode. To further enhance resolution, the electron cascades (produced by the CEMA), are amplified and shaped electronically into uniform square pulses. The shaped signals are detected with an Ithaco 391A lock-in amplifier (LIA). With the aid of a ratiometer feature in the LIA, we are able to measure directly the ratio of the transmission-sputtered current $I_s$ to the incident ion current $I_i$; for $I_i = 1 \mu$A cm$^{-2}$, a ratio of $I_s/I_i$ as small as $1 \times 10^{-9}$ 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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I. 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 (110), (100), and (111) 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 ($\alpha$-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 could result in the transport of SIAs away from the region of a displacement spike$^8$ to produce a depleted (or dilated) 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$^{13}$ have been interpreted as indicating a mean RCS range of 50 Å$^{14}$, 175 Å at 300 K$^{15}$, 1400 Å$^{16}$, 2000 Å$^{17}$, and > 4000 Å$^{18}$. The overall experimental situation for a number of different metals and alloys, with respect to RCSs, has been reviewed by both Venables$^{19}$ and Blewitt et al.$^{20}$

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.$^{21}$ 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 [Fig. 1(a)] or light energetic particles [Fig. 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 \(^{25}\) employing protons that had an incident energy of \(-2.8-3.6\ \text{MeV}\); the protons that emerged from the back surface of the gold films had a mean energy of \(-0.3\ \text{MeV}\).

Figure 1(b) illustrates schematically the physical situation for this type of experiment. The incident protons enter the gold thin film and produce 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 are very spatially extended. \(^{12}\) Both RCSs and FCSs are ejected out of each depleted zone and propagate through the thin film. Those that have a sufficient range, and a large enough energy to overcome the binding forces at the unirradiated surface, produce 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 \((110)\) directions in gold, was greater than 350 eV. Recently, Bay \textit{et al.} \(^{26}\) have studied the transmission sputtering, produced by 500 keV \(\text{Ar}^+\) 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 data fit the Sigmund \(^{27}\) 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 \(^{28}\) 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. Chrens \textit{et al.} \(^{29}\) have studied the transmission sputtering of \((111)\) gold films \((200-1000\ \text{Å thick})\), employing 0.4-1.15 MeV electrons; in this energy range, \textit{no} permanent radiation damage is produced. \(^{30}\) The atoms or ions ejected from the unirradiated surface of the films were collected on a substrate and detected by x-ray microanalysis. \(^{31}\) Ejection patterns were observed, and they exhibited strong three-fold symmetry. For the very low transferred energies involved in this experiment, the ejection patterns must have been caused by FCSs with a range of only one to three interatomic spacings. \(^{29}\)

It is emphasized that the four transmission-sputtering experiments discussed above are of the type illustrated in Fig. 1(b), where the incident irradiating particles pass 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 Fig. 1(a). In this case, the mean range of the low-energy heavy ion is always very small compared with the thickness of the gold thin film, so that all the vacancy type damage is located very close to the irradiated surface. The FCSs and/or RCSs which are ejected from each damaged region (depleted zone) in the forward direction result in ejected particles if they have a range greater than the film thickness, and sufficient energy to overcome the binding forces at the \textit{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 is left behind near the irradiated surface (RCS) or at the \textit{unirradiated} surface (FCS). It should be noted (see Fig. 2) that if the range of some or all the RCSs are less than the thickness of the thin film that SIAs would be deposited within the specimen; furthermore, if the SIAs are mobile at the irradiation temperature, they would cluster to form dislocations loops. \(^{15}\) The only published transmission-sputtering experiment [of the type illustrated in Fig. 1(a)], was performed by Ecker. \(^{14,31}\) He employed both \((100)\) or \((111)\) gold thin films and 0.5 keV \(\text{Ar}^+\), 1 keV \(\text{Kr}^+\), or 0.25-1 keV \(\text{Xe}^+\) ions as the irradiating species; the thicknesses of the gold thin films ranged from 35 to 150 Å. For all of these cases, Ecker stated that the measured transmission-sputtering yields were below the detection limit \([1-5 \times 10^{-3} \text{atom ion}^{-1}]\) of his technique. From these experiments, he concluded that less than \(10^{-2}\) of the FCSs or RCSs produced
had traveled more than 50 Å along (110)-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 to this problem, which employs two channeltron electron-multiplier arrays (CEMAs) in the Chevron configuration as a particle detector; for the xenon energies we employed, our experiment is of the type illustrated in Fig. 1(b). This paper (Part I) presents the apparatus and the measurement technique, while Part II concentrates on experimental results. At the heart of the total system is the Chevron CEMA detector and a lock-in amplifier (LIA) which is 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, is analyzed by a crossed magnetic- and electric-mass analyzer. The gold thin-film specimens can be irradiated anywhere between 25 K and room temperature employing a continuous-transfer liquid-helium cryostat. A transmission electron microscope (TEM) specimen holder is employed to mount each gold film, and after irradiation each film is examined in a TEM to determine its perfection and thickness.

A detailed discussion of the calibration procedure and the experimental errors is given in Secs. III, IV, and Appendix B. 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.

II. EXPERIMENTAL APPARATUS

A. Detection and measuring system

Detection of the transmission-sputtered particles is accomplished dynamically with a Galileo Chevron detector. This (see Fig. 3) consists of two-channel electron-multiplier arrays (CEMAs) in a series; the 37.5-μm-diameter channels within each CEMA are 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 prevents secondary-ion feedback. The front surface of the first CEMA, in our experimental setup, can be biased at a potential anywhere between −1000 to 900 Vdc with respect to the gold thin film; the potential drop across each CEMA is 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, was placed immediately below the Chevron (see Fig. 3). The phosphor screen is maintained at a potential of +2200 Vdc with respect to the bottom surface of the second CEMA when the

![FIG. 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 16.3 Å thick (40 Å in this figure), whereas in practice it was 100-400 Å thick. The incident Xe$^+$ ion beam cannot traverse the thickness of the film, and the Xe ions come to rest within the first layers of the irradiated surface. The maximum energy transferred to a gold lattice atom is 0.96 of the incident ion energy. Within the first few layers, both RCSs and FCSs are produced; for illustrative purposes, we have only indicated the ones which are produced along the [011] and [011] directions. If the range of the FCSs and RCSs is greater than the thickness of film, an atom or ion would be ejected at the unirradiated surface. Note that each RCS leaves behind a vacancy at the unirradiated surface.](image)

![FIG. 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 is observed with the aid of the P-1 phosphor screen that is deposited on the tin oxide-coated glass plate.](image)
A quantity $R$, proportional to $S_{\text{tm}}$, is measured directly by a ratemeter which is built into the LIA. The incident ion current $I$ is first fed into a circuit which measures and averages the ac component of $I$; then a dc voltage $V_I$ proportional to this ac component is fed to the LIA. In addition, the value of $V_I$, which is nearly constant, is measured digitally and recorded by hand. The ratio $R$ of $V_{\text{LIA}} / I$, which is directly proportional to $S_{\text{tm}}$, is measured by the ratemeter. The values of $R$ and $V_{\text{LIA}}$ are recorded as a function of time on a dual-pen strip-chart recorder (see Fig. 4). The conversion of the recorded $R$ values into $S_{\text{tm}}$ values, and the calibration of the system, is discussed in Sec. III.

**B. Low-energy gas-ion gun**

A schematic drawing of the low-energy gas ion gun (herein after called an ion gun) is shown in Fig. 5. The ion gun consists of a plasma chamber at earth potential, a resistively heated tungsten filament, an extractor lens, and a focusing lens: the plasma chamber sits in the center of an axial magnetic field produced by a solenoid. In practice, high-purity (< 20 ppm total impurities) xenon gas is bled into the plasma chamber, via a Varian leak valve, and ionized electrons which are thermionically emitted from the negatively biased (-45 Vdc) tungsten filament. The axial magnetic field is used to increase the electron path length. The Xe$^+$ ion beam is extracted and focused employing lenses which are biased at earth and a negative potential, respectively. In addition, the specimen holder and mass analyzer (see Sec. II C and Fig. 6) are maintained at a negative potential (300 and 700 Vdc); therefore, since the plasma chamber is maintained at earth potential, the ion energy at the specimen is determined by this negative potential. To produce a pulsed Xe$^+$ ion beam, the negative bias on the tungsten filament is pulsed to -45 V at 100 Hz with a duty cycle of 0.5 (see Fig. 3); this provides a pulsed ion current which has 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, is ~1 $\mu$A cm$^{-2}$, while for the 300 eV Xe$^+$ ions, the maximum current density is ~0.4 $\mu$A cm$^{-2}$.

**FIG. 4. A schematic diagram of the electronic circuit employed to measure the transmission-sputtering yields. This circuit is described in detail in Sec. II A.**
C. 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 $S_{im}$ which were $> 10^{-9}$ for films as thick as 800 Å. These signals were most likely due to the channeling of light ion and energetic neutral 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 is analyzed with the mass spectrometer shown in Fig. 6. The ion beam passes through the first and second apertures (0.254 cm in diameter) before it enters the analyzing section of the mass spectrometer. Next, the ion beam passes through a pair of parallel electrostatic deflection plates at a potential $V_p = V'/2$, where $V_p$ is the potential of the ion beam and $V'$ is the potential difference across the plates. Finally, the ion beam passes through a magnetic deflector which consists of a permanent magnet with soft-iron pole pieces$^{44}$: a field of ~2.5 kG was required to deflect a 700 eV Xe⁺ ion beam through 8°. When the mass spectrometer is adjusted for an Xe⁺ ion beam, our design prevents all energetic neutrals and singly charged ions of mass less than 40 amu from reaching the specimen. The analyzed Xe⁺ beam then passes 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 Fig. 6.

In our experiments, the Xe atoms are ionized with 45-eV electrons; at this energy, it is anticipated that ~5% of Xe⁺ ions are doubly charged.$^{40}$ However, an analysis of our mass analyzer indicated that with the electrostatic deflector properly adjusted for Xe⁺ ions, at most 40% of the doubly charged species are able to reach the specimen. Therefore, we expect that < 2% of the xenon ions which actually reach the specimen are doubly charged.

The energy spread of the ion beam was measured for both the 300 and 700 eV Xe⁺ ions. The technique is similar to that used for calibration of the ion current density (see Sec. III); i.e., the thin film specimen was replaced by a small aperture, and the ion current through this aperture was collected and measured, and 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 is $\leq 8$ eV. By this, we mean that the total change in the retarding potential between the point where the measured current begins to decrease, and the point where the measured current goes to zero, is $\leq 8$ Vdc.

D. Total sputtering system

Figure 7 shows the entire sputtering system in its vacuum system. All the components of the sputtering system are suspended from a circular stainless steel plate, for easy de-mounting, which rests on a glass cylinder. The latter in turn rests on a second circular stainless steel plate. The glass cylinder sits inside a fine steel-mesh cylinder which serves as a Faraday cage. The vacuum system is first rough pumped with a vascorb pump, to prevent oil contamination, and then pumped with a 5.08-cm-diameter oil diffusion pump,
III. CALIBRATION AND DATA ANALYSIS

The physical quantity of interest in the transmission-sputtering experiments is \( S_{\text{tm}} \) as a function of the thickness of a gold thin film. To determine \( S_{\text{tm}} \) from the measured quantities \( V_{\text{LIA}} \) and \( I \), we must know \( \lambda_s \) as a function of \( V_{\text{LIA}} \), 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 \( V_{\text{LIA}} \) as a function of \( \lambda_s \) is determined by replacing the sharp electron pulses from the Chevron detector with a pulse generator which operates at a known frequency, and is gated by a square-wave generator operated at the 100-Hz ion-beam frequency. The LIA output \( V_{\text{LIA}} \) as a function of \( \lambda_s \) is found to be linear over the normal operating range for these experiments (\( \sim 1-1000 \) Hz), i.e.,

\[
V_{\text{LIA}} = \kappa \lambda_s, \tag{2}
\]

where \( \lambda_s \) is the measured count rate (number count sec\(^{-1}\)) and \( \kappa \) is a calibration constant.

The quantity \( j \) as a function of \( I \) is 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 \( j \) is given by

\[
j = I' / A_a, \tag{3}
\]

where \( A_a \) is the area of the aperture; the quantity \( j \) is also given by

\[
j = \xi I, \tag{4}
\]

where

\[
\xi = I' / (IA_a) \tag{5}
\]

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 \( S_{\text{tm}} \) defined by Eq. (1) in terms of practical quantities is given by

\[
S_{\text{tm}} = 1.60 \times 10^{-19} \lambda_s / (jA). \tag{6}
\]

Using Eqs. (3) and (4), Eq. (6) becomes

\[
S_{\text{tm}} = 1.60 \times 10^{-19} (V_{\text{LIA}} / I) / (\kappa \xi A), \tag{7}
\]

where the numerical factor converts \( I \) in amperes to number of ions per second.

From the above, it is seen that \( S_{\text{tm}} \) can be determined as a function of time \( t \) by reading the value of the quantity \( R = (V_{\text{LIA}} / I) \) off the strip-chart recorder, and then multiplying by the constant \( 1.60 \times 10^{-19} / (\kappa \xi A) \). The quantity \( S_{\text{tm}} \) increases with \( t \) as the thin film is backspatter-thinned by the \( Xe^+ \) ion beam; the backspattering rate is enormous compared to the transmission-sputtering rate. To determine \( S_{\text{tm}} \) as a function of the film thickness, one must know the total ion dose \( \Phi \) 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 assume that the sputtering rate is not a function of \( \Phi \), although this is not necessarily the case. Even with this assumption, the determination of any of the above quantities presents certain difficulties. As described in

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FIG. 7. The entire sputtering system is 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\(_2\)O\(_3\) (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.

which is trapped with a Granville-Phillips liquid-nitrogen cold trap. The background pressure achieved routinely with this system is \( \sim 10^{-7} \) Torr.\(^{31}\)

The copper specimen-mounting block is cooled by conduction using a continuous-transfer liquid-helium cryostat\(^{32}\) with which the specimen can be maintained at any temperature between \( -25 \) and \( 300 \) K. The entire cryostat is electrically isolated from the copper specimen mounting block by a piece of single-crystal Al\(_2\)O\(_3\) (sapphire). The specimen temperature is measured with a platinum resistance thermometer (item \( H \) in Fig. 7) which is attached to the mounting block. The gold thin film is tightly clamped to the copper specimen holder with the aid of a JEM 200 specimen holder (see Fig. 3). This arrangement guarantees that the gold thin-film strip is at the temperature measured by the platinum resistance thermometer. In Appendix A, we show that the maximum possible temperature rise in the thin film due to ion-beam heating is \( \sim 3 \times 10^{-3} \) K for the geometry employed. The front-surface mirror shown in Fig. 7 is used to observe the pattern produced by the transmission-sputtered particles.
the following paper, the thickness of each thin film is determined by measuring (with 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,\textsuperscript{24} 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 are perfectly uniform, measurement of a nearby section of the same as-deposited film should 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 is found to be better if we measure the final film thickness by TEM and use our own measured value of \( \sim 2.0 \) atom \( \text{ion}^{-1} \) at 700 eV for the backscattering rate (the most commonly used value of energy in our experiments). At 300 eV, our own data is inadequate to determine a backscattering rate, so we assumed that the scaling with bombardment energy was the same as for the published data\textsuperscript{25}; this procedure yields a value of \( \sim 1 \) atom \( \text{ion}^{-1} \) at 300 eV.

**IV. EXPERIMENTAL ERRORS**

The two major sources of measurement error in our experiments are fluctuations in \( I \) and statistical scatter. The former error is peculiar to our experimental setup, while the latter one is intrinsic to the experiment. In Sec. III, we discussed the relationship between \( I \) and \( J \), and showed that if \( J \) is uniform, then the calibration constant \( \xi \) should be independent of operating conditions; unfortunately, it is not uniform. Small changes in the ion-gun operating conditions change \( \xi \) by as much as 30%, although 10% changes are more typical. The changes in \( \xi \) cause similar changes in the output ratio \( R \).

In Appendix B, we present the derivation of an equation for the variance of \( V_{\text{LIA}} \) [\( \text{var}[V_{\text{LIA}}] = \sigma^2 \)] under the assumption that \( V_{\text{LIA}} \) and the noise are both distributed according to a Poisson distribution. We show (see Appendix B) that the ratio of \( [\sigma_v/E_v(V_{\text{LIA}})] \) is given by

\[
\frac{\sigma_v}{E_v(V_{\text{LIA}})} = \frac{1}{\sqrt{RC}} \left( \frac{1}{\lambda_1} + \frac{2\lambda_2}{\lambda_2^2} \right)^{-\frac{1}{2}},
\]

where \( E_v(V_{\text{LIA}}) \) is the expected value of \( V_{\text{LIA}} \), \( R \) and \( C \) are the resistance and capacitance value of the RC filter\textsuperscript{27} in the LIA output section, and \( \lambda_1 \) and \( \lambda_2 \) are the mean pulse frequencies of the signal and noise, respectively. As expected, \( \sigma_v \) is inversely proportional to the square root of RC (i.e., \( \tau_L \)). Also, if \( \lambda_1 > \lambda_2 \), then the noise term can be neglected, but if \( \lambda_1 < \lambda_2 \), then the statistical scatter term dominates \( \sigma_v \).

The quantity \( \tau_L \) is a variable; therefore we could have, in principle, reduced \( \sigma_v \) to any arbitrary extent by increasing \( \tau_L \) (i.e., quantity RC). This approach has several drawbacks. First, since we wish to measure an output as a function of film thickness as the film is sputter-thinned, or equivalently as a function of \( t \), the quantity \( \tau_L \) has to be maintained at a value much less than any characteristic time involved in the change in the output. Second, a large value of \( \tau_L \) implies a long transient response time when the Xe\textsuperscript{+} ion beam is turned on. Because of problems in experiments we had performed earlier (connected with synchronous background signals unrelated to transmission sputtering), the Xe\textsuperscript{+} ion beam is 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 an irradiation is reinitiated, a good transient response (i.e., small \( \tau_L \)) is found to be essential. A third problem with a large value of \( \tau_L \) involves 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 is not perfect, and bursts of noise from nearby laboratories occasionally intruded. The presence of these noise bursts is easily detected with an auxiliary auditory system, and is noted on the strip chart recording of the output. The noise bursts are typically more intense than the real output, and they are capable of causing considerable deviations in \( V_{\text{LIA}} \). For small values of \( \tau_L \), it is practical to ignore the data a few \( \tau_L \) after the noise burst. As \( \tau_L \) increases, more and more data can be lost in this way. In most experiments, a \( \tau_L \) value of either 25 or 80 sec was used, although in a few experiments, a \( \tau_L \) of 250 sec proved necessary; the largest value of \( \tau_L \) did cause difficulties—which will be discussed in Part II.\textsuperscript{35}

**V. SUMMARY**

A novel transmission-sputtering system is described. It has 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, are detected dynamically with a Galileo Chevron detector. When the Chevron detector is operated in the saturated mode, individual transmission-sputtering particles produce sharp electron pulses. In addition, the transmission-sputtering pattern produced is observed with the aid of a phosphor screen placed directly behind the Chevron detector.

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

3. The LIA had a ratiometer feature which allows the direct measurement of the ratio of the transmission-sputtering current \( I_s \) to the incident ion current \( I_0 \); for \( I_0 = 1 \) \( \mu A \) cm\(^{-2} \), a ratio of \( I_s/I_0 \) as small as \( 10^{-9} \) has been measured.

4. The Xe\textsuperscript{+} ion beam used for the sputtering experiments is mass analyzed in a crossed electric- and magnetic-field analyzer. With the mass spectrometer adjusted for an Xe\textsuperscript{+} ion beam, our design prevents 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 indicates that <2% of the xenon ions which actually reach the specimens are doubly charged.

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

(6) The calibration and data analysis procedures are given, and a detailed analysis of the experimental errors involved is presented.

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APPENDIX A: HEATING OF THE SPECIMEN DURING THE ION IRRADIATION

The irradiated portion of the gold thin film has the geometry of a small right-circular cylinder with a radius $r_0$ that is either 25 or 50 $\mu$m and a height $l_0$ that ranges from $10^{-8}$ to $4 \times 10^{-6}$ cm. This cylindrically shaped volume is situated in the center of a strip of gold thin film that is ~0.1 and ~0.3 cm on each side. The surrounding strip of gold thin film is so well clamped in the specimen holder that we assume that its temperature is unaffected by the irradiation (see Fig. 3). The governing steady-state heat equation for this problem is

$$\frac{1}{r} \frac{d}{dr} \left( r \frac{dT}{dr} \right) + \frac{Q}{l_0 K} = 0,$$

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 $r_0/l_0$ is always much greater than unity so we treat the heat-flow problem as a one-dimensional one in the plane of the thin film. The boundary conditions used are: (1) the temperature is a constant at $r = r_0$ and equal to the temperature measured by the platinum resistance thermometer; and (2) the slope (dT/dr) at $r = 0$ is equal to zero. The integration of Eq. (A1) twice and the evaluation of the constants of integration employing the above boundary conditions yields the following expression for the maximum temperature rise:

$$\Delta T_{\text{max}} = \frac{r_0^2}{4l_0 K} Q.$$

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

APPENDIX B: ERROR ANALYSIS OF THE STATISTICAL SCATTER IN THE MEASURED TRANSMISSION-SPUTTERING YIELD ($S_{\text{um}}$)

To calculate the expected statistical scatter in the measured values of $S_{\text{um}}$ (see Sec. III) we employ a simple model for the LIA. As shown in Fig. 8, we take the LIA to be simply a voltage inverter with a switch and an RC filter; the switch is controlled by the ion-beam pulser. When the Xe$^+$ ion beam is on, the signal passes directly to the RC filter. Alternatively, when the Xe$^+$ ion beam is off, the signal is inverted. For the purpose of the calculation, this configuration is employed to represent an ideal LIA.

The Xe$^+$ ion beam is pulsed as shown in Fig. 9(a). Each ion-beam pulse initiates a number of randomly distributed signal pulses, as indicated schematically in Fig. 9(b). In addition to the signal pulses, there are a random distribution of noise pulses, as shown in Fig. 9(c). The sum of the signal plus the noise pulses, through the inverter and switch, gives the voltage pulses $V_t$ exhibited in Fig. 9(d).

The signal and noise pulse that reach the LIA are all identical, and their time duration is short compared to the RC time constant $\tau_R$ of the LIA. Therefore we ignore the actual shape of the signal and noise pulses and replace them by delta functions; that is, a single pulse $P(t_0)$ at time $t_0$ is represented by

$$P(t_0) = P_0 \delta(t - t_0),$$

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 $V_{\text{in}}$ is represented by a steady stream of random pulses $S(t)$ modulated by a function $g(t)$; therefore, $V_{\text{in}}$ is represented by

$$V_{\text{in}} = g(t) S(t),$$

where

$$S(t) = \sum_{n} P_0 \delta(t - T_n).$$

Also,

$$g(t) = \begin{cases} 1 & 0 < t < (n + 1)T \\ 0 & (n + 1)T < t < (n + 1)T \end{cases}$$

FIG. 8. The schematic model of the lock-in amplifier employed to calculate the expected statistical scatter in the transmission-sputtering yield.
where \( \lambda_s \) and \( \lambda_n \) are the mean pulse and signal frequencies for the signal and noise, respectively. The expectation value of \( V_{\text{LIA}}(t) \) is then given by

\[
E[V_{\text{LIA}}(t)] = \int_{-\infty}^{\alpha T} e^{-\alpha t - \gamma}[g(\tau)E[S(\tau)] + m(\tau)E[N(\tau)]]d\tau
\]

\[
= \int_{-\infty}^{\alpha T} e^{-\alpha t - \gamma}[P_{\alpha L_s}g(\tau) + P_{\alpha L_n}m(\tau)]d\tau
\]

\[
= E_s[V_{\text{LIA}}(t)] + E_n[V_{\text{LIA}}(t)], \quad (B12)
\]

where

\[
E_s[V_{\text{LIA}}(t)] = P_{\alpha L_s} \int_{-\infty}^{\alpha T} e^{-\alpha t - \gamma}g(\tau)d\tau \quad (B13a)
\]

and

\[
E_n[V_{\text{LIA}}(t)] = P_{\alpha L_n} \int_{-\infty}^{\alpha T} e^{-\alpha t - \gamma}d\tau. \quad (B13b)
\]

The integrals in Eqs. (B13) can be evaluated in a similar fashion. First, consider Eq. (B13a) for the interval \( nT < t < (n + 1)T \), it is given by

\[
E_s[V_{\text{LIA}}(t)] = P_{\alpha L_s} e^{-\alpha t} \left( \int_{nT}^{(n+1)T} e^{\alpha \tau}d\tau + \text{higher order terms} \right)
\]

\[
= P_{\alpha L_s} e^{-\alpha t} \left( \int_{nT}^{(n+1)T} e^{\alpha \tau}d\tau + \sum_{k=1}^{\infty} \int_{(n-k)T}^{(n-k+1)T} e^{\alpha \tau}d\tau \right). \quad (B14)
\]

Equation (B14) is constructed with the aid of Fig. 9a by considering the time \( t \) between \( nT \) and \( (n + 1)T \). Equation (B14) is evaluated with the aid of the sum

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

to yield

\[
E_s[V_{\text{LIA}}(t)] = \frac{P_{\alpha L_s}}{\alpha} \left[ 1 - e^{\alpha T - t} \left( \frac{1 - e^{-\alpha T/2}}{1 - e^{-\alpha T/2}} \right) \right]. \quad (B15)
\]

In a similar manner, we obtain

\[
E_n[V_{\text{LIA}}(t)] = \frac{P_{\alpha L_n}}{\alpha} \left[ 1 - 2e^{\alpha T - t} \left( \frac{1 - e^{-\alpha T/2}}{1 - e^{-\alpha T/2}} \right) \right] \quad (B16)
\]

where the factor of two difference is due to Eqs. (B7). Since the quantity \( \alpha T \) is \( \ll 1 \), Eqs. (B15) and (B16) reduce to

\[
E_s[V_{\text{LIA}}(t)] \approx P_{\alpha L_s}/(2\alpha), \quad (B17)
\]

and

\[
E_n[V_{\text{LIA}}(t)] \approx P_{\alpha L_n}(t - nT). \quad (B18)
\]

It is clear that since \( 1/(2\alpha) \) is large compared to \( (t - nT) \), \( E_n[V_{\text{LIA}}(t)] \) is \( \ll E_s[V_{\text{LIA}}(t)] \). Employing Eq. (B17), we experimentally determine \( P_{\alpha L} \) by measuring \( V_{\text{LIA}} \) for a known \( \lambda_s \).

The variance of \( V_{\text{LIA}} \) [\( \text{var}(V_{\text{LIA}}) = \sigma^2 \)] is given by the usual relationship
\[
\text{var}[V_{LIA}] = E[V_{LIA}^2] - [E[V_{LIA}]]^2. \tag{B19}
\]

The quantities \(V_{LIA}^2(t)\) and \(E[V_{LIA}^2]\) are given by

\[
V_{LIA}^2(t) = \int_\tau^\infty \int_v^\infty \text{d}v e^{-\alpha t - \tau} e^{-\alpha v - v} \times \left[ g(\tau) S(\tau) + m(\tau) N(\tau) \right] \cdot [g(v) S(v) + m(v) N(v)], \tag{B20}
\]

and

\[
E[V_{LIA}^2(t)] = \int_\tau^\infty \int_v^\infty \text{d}v e^{-\alpha (t - \tau) - \alpha (v - v)} \times \left[ g(\tau) S(\tau) + m(\tau) N(\tau) \right] \cdot [g(v) S(v) + m(v) N(v)] \tag{B21}
\]

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

\[
E[S(\tau)N(\tau)] = E[S(\tau)]E[N(\tau)] = P^2_0 \lambda_s \lambda_a. \tag{B22a}
\]

Also, it is known that the two auto-correlations \(E[S(\tau)S(\tau)]\) and \(E[N(\tau)N(\tau)]\) are given by

\[
E[S(\tau)S(\tau)] = P^2_0 \lambda_s^2 + \lambda_s P_0^2 \delta(\tau - v), \tag{B22b}
\]

and

\[
E[N(\tau)N(\tau)] = P^2_0 \lambda_a^2 + \lambda_a P_0^2 \delta(\tau - v). \tag{B22c}
\]

Therefore, Eq. (B21) becomes

\[
E[V_{LIA}^2(t)] = \int_\tau^\infty \int_v^\infty \text{d}v e^{-\alpha (t - \tau) - \alpha (v - v)} \times \left[ \lambda_s P_0^2 g(\tau) S(\tau) \right] \cdot [g(v) S(v) + m(v) N(v)] + \lambda_a P_0^2 m(\tau) N(\tau) \tag{B23}
\]

Comparing Eq. (B23) with Eq. (B12), it is clear that

\[
E[V_{LIA}^2(t)] = \left\{ E[V_{LIA}^2(t)] \right\}^2 + \int_\tau^\infty \int_v^\infty \text{d}v e^{-\alpha (t - \tau) - \alpha (v - v)} \times \left[ \lambda_s P_0^2 g(\tau) S(\tau) \right] \cdot [g(v) S(v) + m(v) N(v)] + \lambda_a P_0^2 m(\tau) N(\tau) \tag{B24}
\]

Finally, from (B19), and integrating over \(v\), we obtain

\[
\text{var}[V_{LIA}](t) = \int_\tau^\infty \text{d}t e^{-2\alpha (t - \tau)} \cdot \left[ \lambda_s P_0^2 [g(\tau)]^2 \right] + \lambda_a P_0^2 \cdot \left[ m(\tau) \right]^2, \tag{B25}
\]

employing

\[
[g(\tau)]^2 = g(t) \tag{B26a}
\]

and

\[
[m(\tau)]^2 = 1. \tag{B26b}
\]

[See Eqs. (B4) and (B7) to understand Eq. (B26a).] The integration of Eq. (B25), employing Eqs. (B15) and (B26), yields

\[
\text{var}[V_{LIA}](t) = \frac{\lambda_s P_0^2}{2\alpha} \cdot \left[ 1 - e^{-2\alpha (t - \tau)} \right] \cdot \left[ \frac{1 - e^{-\alpha t}}{1 - e^{-\alpha T}} \right] + \lambda_a P_0^2 \cdot \frac{2\alpha}{\alpha} \tag{B27}
\]

Since \(\alpha T \ll 1\), Eq. (B27) reduces to

\[
\text{var}[V_{LIA}](t) = \frac{\lambda_s P_0^2}{4\alpha} + \frac{\lambda_a P_0^2}{2\alpha}. \tag{B28}
\]

Finally, employing Eq. (B17), we obtain for the expected relative deviation in \(V_{LIA}\)

\[
\frac{\text{var}[V_{LIA}]}{E[V_{LIA}]} \approx \frac{\lambda_s}{\lambda_s^2 + 2\lambda_a} \approx \frac{1}{\lambda_s \lambda_a} \tag{B29}
\]


We restrict our discussion to gold since this metal has been studied extensively, and yet the results have remained controversial for more than a decade.


The reader is referred to Thompson,22, Nelson,23, and Carter and Collignon24 for further details and results concerning backscattering experiments.


For the Cherns et al.29 experiment, no radiation damage was produced; therefore, when thinking about this experiment, the damaged regions (de-
pleted zones) must be mentally omitted.


Galileo Electro-Optics Corp., Technical Memorandum 600 (Galileo Park, Sturbridge, Massachusetts, 1972).


G. Ayrault and D. N. Seidman, Cornell Materials Science Center Report No. 2945 (1982).

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


In the case where negatively charged ejected species are detected (see Sec. 3.4 of Part II), these species also have, 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. 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.

The value of $V_{s}$ was proportional to the in-phase Chevron count rate averaged over a time period $\tau$, of 12.5-125 sec.

The present ion gun was designed on the basis of several earlier designs reported in the literature.


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, is 17°.


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


Backscattering yield data are available for Xe$^+$ on polycrystalline Au for bombardment energies up to 600 eV. An extrapolation of this data to 700 eV yielded a value of $\sim 3.7$ atom ion$^{-1}$. This value was obtained for very high Xe$^+$ doses on Au and for polycrystalline specimens. Thus we attribute the difference between our value of the backscattering rate and the Rosenberg and Wehner value to difference in the experimental conditions.
