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New Method for Measuring Sputtering in the Region Near Threshold

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A new method is described by which a plated quartz crystal oscillator is used for the measurement of sputtering. The crystal is placed in a molecular beam and sputtering of its plating is measured by the frequency change of the oscillator. The relationship between the frequency change of the oscillator and the change in plating mass of the crystal is given. The method is extremely sensitive. Sputtering of less than a millimicrogram of plating can be measured. A wide range of material can be used to plate the crystal. A crystal oscillator sputtering gauge was built by using a frequency counter with a digital readout to record the oscillator frequency. The operation of the gauge is illustrated by preliminary measurements on the sputtering rates of gold in an argon beam between 0 and 100 ev. Other uses of the gauge, such as a neutral beam detector, are also discussed.

INTRODUCTION

THE study of sputtering of surfaces by high velocity molecules has taken on new importance with the advent of satellites and space vehicles. The particles of the upper atmosphere and space impact on the surface of a vehicle with a high velocity. If the velocity is above the threshold of sputtering, the surface of the vehicle will be slowly eroded. Over a period of time, depending on the sputtering rate, the original values of surface properties such as drag coefficient and reflectivity will change. At the present time it is difficult to tell how these changes will affect the useful life of the vehicle because practically no measurements of sputtering rates have been made on surface materials in the region of threshold.

The reason for the lack of measurements is the inadequacy of conventional methods to measure in a short

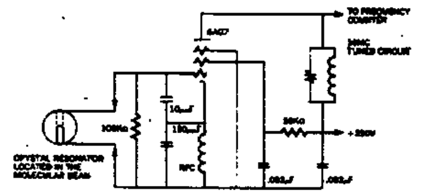

FIG. 1. 10-Mc crystal oscillator.

period the vanishingly small amounts of material sputtered. In general the methods are all complex and time consuming. In particular, determinations which employ densitometer techniques or those in which the sputtered material is actually weighed are not sensitive enough. Methods of high sensitivity such as the use of tracer atoms[1] or surface ionization[2] for detection are not the answer either because they can be used with relatively few materials. A new spectroscopic method has been reported which can be used with all materials.[3] It appears to be very useful in the detection of sputtering but since the method is indirect it is difficult to tell whether or not measurements made at low energies will be reliable. To overcome the drawbacks of available methods a new method has been developed by which a plated quartz crystal oscillator is used for the measurement of sputtering. The method is highly sensitive, versatile, and direct. The crystal is placed in the molecular beam and sputtering of its plating is measured by the frequency change of the oscillator. It is the purpose of this paper to describe a sputtering gauge which was built using such an oscillator.

DESCRIPTION AND ANALYSIS OF THE GAUGE

The gauge consists of a 10-Mc plated optically polished AT cut quartz crystal, an electronic oscillator for driving the crystal, and a frequency counter.

Figure 1 is the circuit diagram[4] of the electronic oscillator. Its frequency is critically dependent on the plating mass of the crystal which is only a few micrograms. Removal of less than a millimicrogram is enough to change the oscillator frequency a measurable amount.

An energized crystal is a mechanical vibrating plate which is coupled to the driving oscillator by the piezoelectric effect. Its electronic frequency is dependent on its equivalent electrical parameters.[5] The parameter of importance in the operation of the gauge is its equivalent inductance which is dependent on the resonating mass of the crystal including its plating. Equation (1) expresses the frequency, in cycles per second, in terms of the crystal thickness[6] t, in millimeters. Using the equation one can compute the frequency change Δf produced by a removal of a thickness Δt of the plating. It is assumed that the thin plating behaves like an equal mass of quartz. The constant k is characteristic of quartz and is independent of the plating.

$$f=k/t, \tag{1}$$

where k equals 1.66×10^6 cycles-mm/sec.

$$\Delta f=-k\Delta t/t^2. \tag{2}$$

By using a precision 10-Mc crystal, the plating thickness removed for a measurable 1-cycle frequency increase is the following:

$$\Delta t=-kf/f^2=-1.66\times10^{-8}\ \text{mm}=-0.166\ \text{angstrom}. \tag{3}$$

Since Δt is inversely proportional to f^2, a higher fundamental frequency would give even greater sensitivity. For example by using a 20-Mc crystal, the highest available commercially, an average thickness change of only 0.04 A could be measured.

By using a circular plating 4 mm in diameter and assuming the motional area A is of the same dimensions which is reasonable for a 10-Mc crystal, the following decrease Δm in plating mass could be measured:

$$\Delta m=\rho A\Delta t, \tag{4}$$

$\Delta m=-5.5\times10^{-10}$ grams where ρ is the density of quartz, 2.6 g/cm^3. It should be noted from Eqs. (2) and (4) that an equal mass deposited on the crystal would produce a frequency decrease. Hence, the crystal can be also used to detect deposition of extremely small amounts of material on its motional surface. For example, the gauge could be

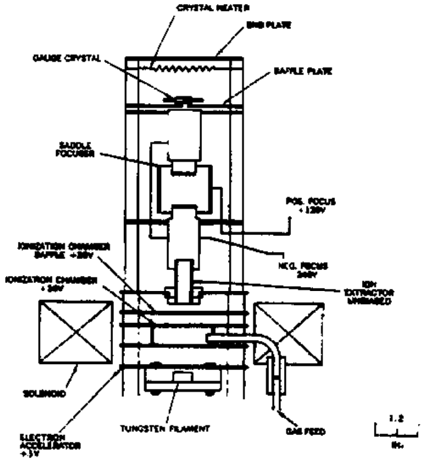

FIG. 2. Beam generator assembly.

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[1] N. D. Morgulis and V. D. Tishchenko, Soviet Phys.—JETP 3, 1 (1956).

[2] R. C. Bradley, Phys. Rev. 93, 719 (1954).

[3] R. V. Stuart and G. K. Wehner, Phys. Rev. Letters 4, 409 (1960).

[4] *The Radio Handbook* (Editors and Engineers Limited, Summerland, California, 1959), 15th ed., p. 248.

[5] D. McKeown, Proceedings of the Eleventh Annual Frequency Control Symposium, Fort Monmouth, New Jersey, 1958, p. 317.

[6] W. P. Mason, *Piezoelectric Crystals and their Applications to Ultrasonics* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1956), p. 55.

used to measure sputtering indirectly by collecting on the surface of a crystal the material sputtered from a target.

EXPERIMENTAL SETUP

Figure 2 is a drawing of the beam generator assembly. It consists of an ion source, focuser, and target chamber. A Finkelstein[7] type ion source was used. The ions are extracted from the source and focused by the saddle focuser. They then pass through a $\frac{1}{8}$-in. diam hole in a baffle plate into the target chamber. The crystal is mounted in this chamber and heated to 60°C to keep any mercury present in the vacuum chamber from collecting on it. Higher temperatures were not required because when the beam was turned off the frequency of the crystal did not change a significant amount over a long period of time. At lower temperatures the frequency decreased a few cycles per minute indicating that mercury was collecting on the plating of the crystal.

The assembly was set up in a glass chamber evacuated by a mercury diffusion pump. Two liquid nitrogen cold traps isolated the pump from the chamber. The pressure in the chamber reached 1×10^{-7} mm Hg with the beam off and did not rise above 2×10^{-4} mm Hg with the beam on.

Figure 3 is a plot of the energy in an argon (Ar^+) beam generated by the ion source. On using 30-ev electrons to ionize the argon no significant amount of Ar^{++} is produced. The ion source was biased at 30 v above ground so that the beam striking a target for example at ground potential had an energy of approximately 30 ev. The plot was obtained then, by biasing the target between +30 and −70 v. At a beam intensity of 3×10^{-6} amp cm^{-2} the background pressure in the vicinity of the target was 1×10^{-4} mm Hg.

RESULTS

Figure 4 shows the sputtering rates, as a function of frequency change of the gauge, for a thin gold plating in a normally incident Ar^+ beam. Curve A was taken at 1.5 and curve B at 1.0 μa. Even with beams in the microampere range each point on the curve could be taken after only 30 sec of sputtering because the sensitivity of the gauge was high. Temperature cut crystals whose frequency changed less than 0.0001%/°C were used and their temperature controlled to 0.1°C by an oven. The energy transferred to the crystal by the beam had no significant effect on its frequency. The error in frequency, due to temperature variations of the crystal, is ±0.5 cycle. The energy given for every point is the maximum energy of the beam.

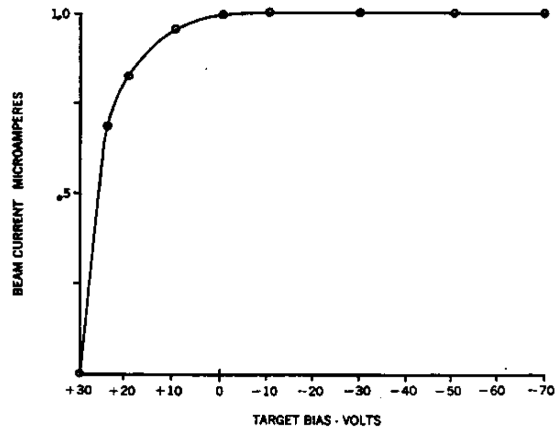

FIG. 3. The beam current as a function of target voltage.

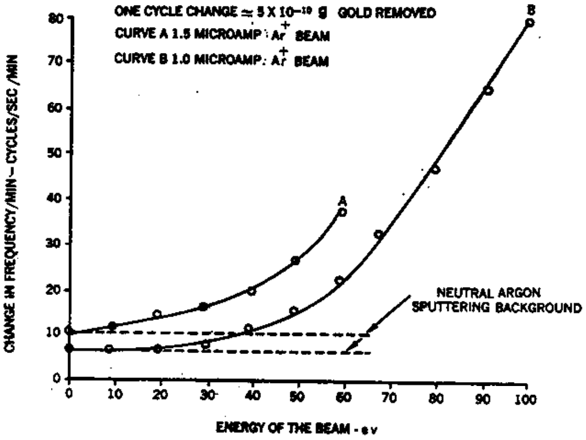

FIG. 4. Crystal frequency change as a function of beam energy.

Present in the ion beam at constant intensity and independent of its energy, was a 0- to 270-ev neutral beam. It was generated in the extractor and first stage of the saddle-focuser by a charge exchange process. The success of the sputtering threshold measurements was due to this neutral beam present in the ion beam. It cleaned from the surface of the crystal the minute quantities of dirt which built up on it when it was bombarded by ion beams below 60 ev. By baking the vacuum chamber for long periods the amount of this material could be reduced but not eliminated. Runs which were made without the neutral beam indicated that sputtering ceased at 60 ev. Later measurements showed this to be not the threshold of gold but of the deposit covering it. Figure 5 is a crystal typical of those used in the gauge bombarded for several hours by a 50-ev beam showing the deposit. The long period was required before it became visual. Afterward the deposit was partially removed by a 300-ev ion beam.

The presence of neutrals and the deposition of impurities on the crystal was found because of a very important feature of the gauge. It operates while in the beam. If the gauge frequency is increasing, a net sputtering reaction is occurring. By biasing the crystal to repel the ions, the gauge detects the neutral component of the beam whose energy is above the sputtering threshold. If the gauge fre-

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[7] A. T. Finkelstein, Rev. Sci. Instr. 11, 52 (1940).

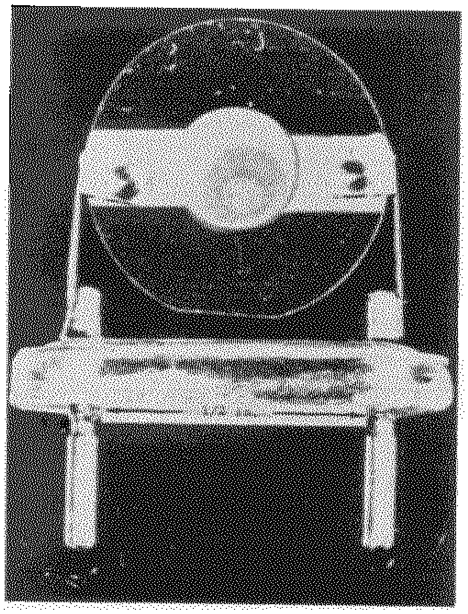

FIG. 5. 10-Mc crystal unit showing impurities deposit.

quency decreases, a net deposition is occurring on the crystal. The gauge can then be used to detect neutral beams which stick to the crystal or chemisorption in beams which react with the plating of the crystal.

By making use of Eq. (4) which relates the frequency change of the crystal to the mass removed and subtracting the effect of the neutral beam in Fig. 4, Fig. 6 was plotted. Only values are used from the 1.5-μa beam below 40 ev because the low sputtering rates of the 1.0-μa beam were partially masked by the deposition of the impurities on the crystal. The curve shows μ, the sputtering rate in atoms per ion, as a function of beam energy. The magnitude of μ may be low by about 20% because γ, the ratio of secondary electrons to incident ions, is unknown and may be as large[8] as 0.2. The values of μ between 100 and 50 ev agree well with Wehner's results,[8] the crosses along the curve, and show that the sputtering rates of thin gold films are close to those of polycrystalline used by him. Appropriate measurements are planned to confirm this.

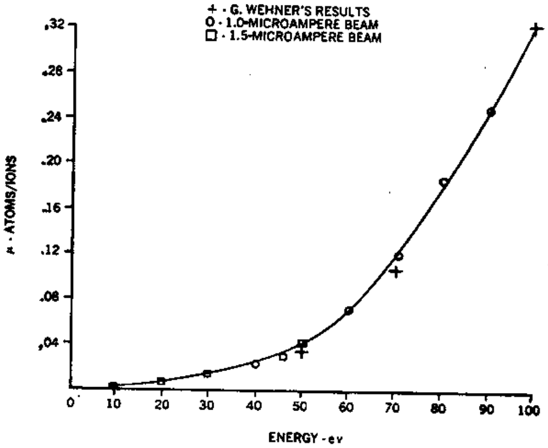

FIG. 6. Sputtering rates of gold in argon at normal incidence.

The threshold found here of about 10 ev is considerably lower than that of 50 ev found by Wehner. It would be unwise though to accept a lower threshold on the basis of these measurements because of the presence of the high neutral beam sputtering background. More measurements without the neutral beam are being planned now that the gauge is perfected.

ACKNOWLEDGMENTS

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[8] G. K. Wehner, Conference Report, The Rand Corporation Conference on Aerodynamics of the Upper Atmosphere (1959, unpublished).