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STUDY OF LASER DEPOSITED THIN FILMS

By James A. R. Samson

GCA CORPORATION

Bedford, Massachusetts

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SUMMARY

This is the final report on the "Study of Laser Deposited Thin Films" performed under Contract No. NAS5-11033. The aim of this program has been to investigate the feasibility of using a pulsed laser beam as a heat source to evaporate metals for the production of high quality mirrors. In the course of this work it has been shown that the laser will evaporate all types of metals including the refractory types. Mirrors of Al, W, Mo, etc., have been produced. It was known that considerable "sputtering" of particulate material was produced when the metal was vaporized by a laser beam. This "sputtering" decreased the usefulness of the resulting mirrors. The program was thus aimed at understanding the mechanism of the unwanted "sputtering" in the hope that this would lead to the production of high quality mirrors. Considerable success has been achieved in this direction.

INTRODUCTION

The evaporation of metals by a laser beam has been investigated by several groups. In some cases, the object was to produce an intense source of ions and electrons; in other cases, the evaporated material was used in mass analysis of surfaces. However, no detailed study exists of the production of high quality reflectance surfaces for vacuum uv radiation. It is the purpose of this contract to study the feasibility of producing high quality mirrors. It has already been established that a ruby laser (50 to 100 joules), operating in the normal mode, can evaporate material from most metals, including such refractory metals as tungsten and iridium. However, in addition to vaporized material being emitted from a target, considerable "sputtered" material is also ejected. "Sputtering" as used in this report is defined as the ejection of globules of particulate matter. Since these globules of metal are sources of scattered light, they must be eliminated in order to obtain good quality mirrors. The angular distribution of this "sputtered" material was investigated, and the design of targets which might reduce the amount of "sputtering" is discussed. In this report, the use of these targets and their effectiveness in reducing "sputtering" is described. Other deleterious effects and their cure are discussed. The greatest success has been the use of powdered metals compressed in "pill" form. It appears as if the reduced heat conductivity of the metals in this form decreases the amount of "sputtering."

A glass evaporator system was used to evaporate and prepare mirrors of tungsten and molybdenum. These mirrors were not entirely free from "sputtered" material. However, the molybdenum mirror was excellent and reflectance measurements were performed and are presented in this report.

This report describes the apparatus and the techniques used to minimize "sputtering." The initial aim of this study was to determine the angular distribution of the "sputtered" material and endeavor to understand the production mechanism. From this better understanding, it was hoped to reduce or eliminate the "sputtering." Although not required by the work statement, the results of a literature search for appropriate discussions on laser evaporations of metals is included. The list of papers with their titles are given at the end of the report under General Bibliography.

DISCUSSION

The ruby laser used for this program is a Korad Model K2-QP which can be used in two modes. The following are the typical specifications:

Normal Mode

Maximum Energy Output	= 100 joules
Pulse Width	= 500 μsec
Wavelength	= 6943 Å
Line Width	= 0.05 Å
Beam Divergence	= 5 milliradians
Beam Size at Exit	= 1.9 cm
Degree of Polarization (Electric Vector)	= 100 percent in horizontal plane

Q-Switch Mode

Energy Output	= 1 to 10 joules
Peak Power (Max.)	= 750 megawatts
Pulse Width	= 10 nanoseconds
Polarization - as above	

The high vacuum pumping system was made from pyrex. A three-stage glass diffusion pump was used with a liquid nitrogen cold trap. The evaporation chamber was constructed from a 500 mℓ flask with the target materials located at the center of the spherical bulb. The reason for this was to determine the spatial distribution of the evaporated material relative to the axis of the laser beam. The evaporated material was simply allowed to coat the walls of the spherical flasks. Figure 1 shows the experimental arrangement of the laser and 500 mℓ flask. The additional side arm holding the substrate was added at a later date.

A lens of 100 mm focal length was used to focus the laser beam onto the sample face. The spot diameter, d, of the light beam at the focal plane of a lens is given by

$$d = 2f\theta \qquad (1)$$

where θ is the beam angle insofar as the lens aberrations can be neglected. For this laser $\theta \approx 0.005$ radians. Thus, d = 1 mm.

Results

The laser beam was fired through the base of the 500 mℓ flask which was subsequently coated by the evaporated material. Since the target was located at the center of the sphere, the coated surface gave a true representation of the angular distribution of the vaporized and sputtered material. The surprising result was that the sputtered particles tend to

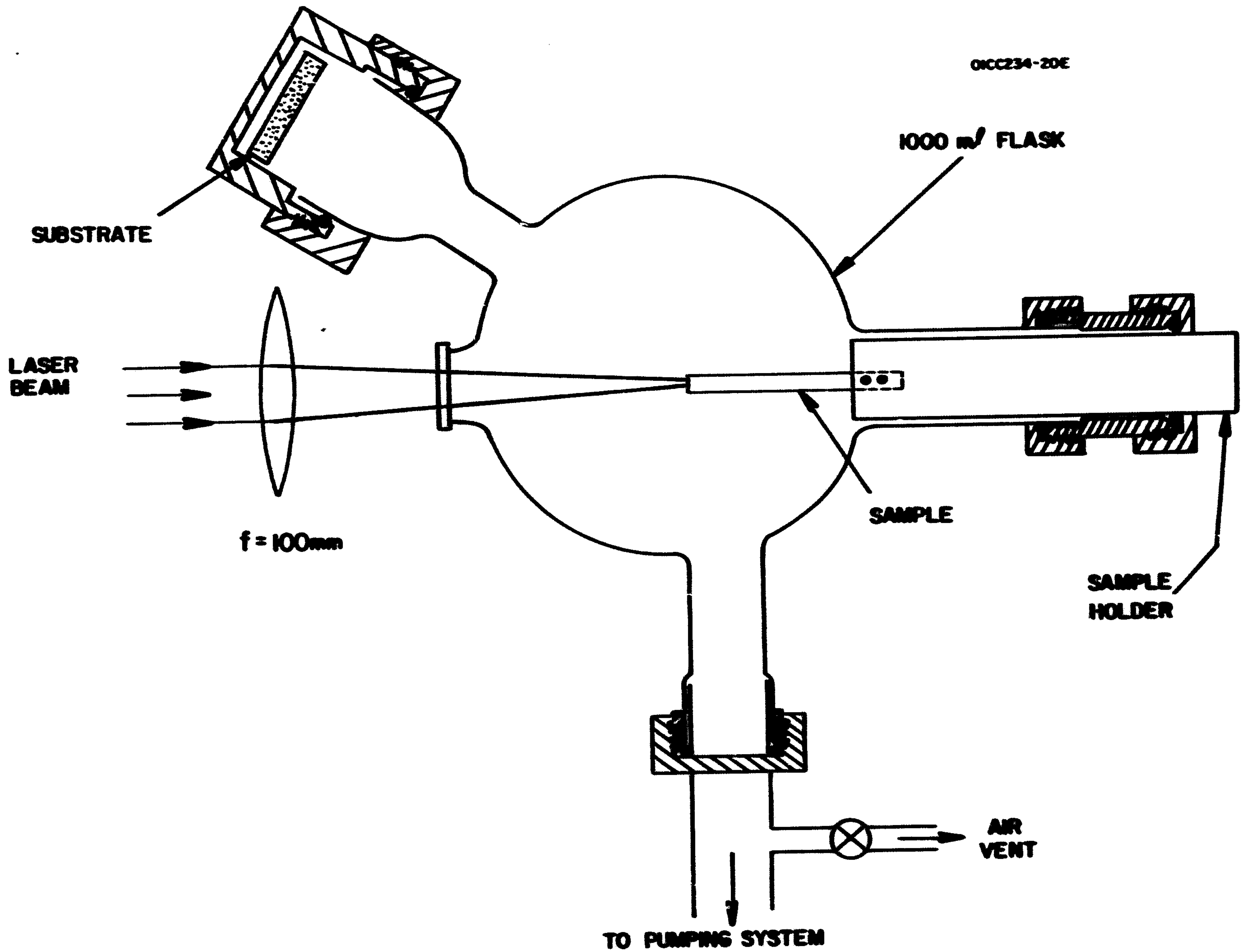

Figure 1. High vacuum evaporator.

fly off at angles greater than ±30 deg. from the axis of the beam, whereas the evaporated material tended to be projected in the direction of the beam with a spatial distribution which varied approximately as cos θ or cos θ^2, where θ is the angle with respect to the beam. Figure 2 indicates the approximate condition of the hemisphere. The more densely sputtered areas tended to lie in the horizontal plane. The clear coating still contained some "sputtered" material. The basic condition shown in Figure 2 was reproducible at laser power levels of 30, 56, and 72 joules. With the lower power levels, less evaporation per pulse resulted. At the first two power levels, two shots were required to produce a significant coating, whereas at 72 joules, one shot was sufficient. In all cases, the end of the target sample faced the laser beam at right angles and had dimensions of 3 x 3 mm. The following model is suggested to explain the results:

Referring to Figure 3, it would appear that at the instant of impact, the laser beam would cause an area of vaporized metal with a cooler liquid area surrounding the vapor. As the vapor expands in all direction (deton ation), the molten metal is squeezed out of the annular region surrounded by solid metal and tends to fly off at angles greater than $\theta = 0$. The vaporized region is formed within the central cone and is ejected in a directional fashion about the beam axis.

A laser pulse operating in the normal mode contains of the order of 500 spikes per pulse, each pulse lasting for about 500 usec. Thus if the heat conduction of the metal is slow compared to times of the order of microseconds, each spike heats the metal until it liquifies. Subsequent spikes act as projectiles striking the liquid metal and sputtering material in much the same manner as a liquid drop landing on a liquid surface. Figure 4 illustrates the splash obtained by a milk drop landing on a shallow pool of milk. The drawing is a reproduction from a photograph obtained by Prof. Edgerton using flash photography. The figure illustrates the fact that the splash creates globules of the liquid and ejects them into a cone. It is suggested that the laser pulse is analogous to the liquid drop model. If this is indeed the case it would be desirable to reduce or remove the liquid pool stage or to use only a single spike in the laser pulse as in the Q-mode of operation. With the present laser the total energy output in the Q-switched mode is about one-hundred times less than in the normal mode and no evaporation was observed. One could compensate for this lower overall energy by using a shorter focal length lens producing an image 0.1 mm in diameter rather than the present 1-mm diameter image. Therefore, the energy density would be about one-hundred times greater. However, this would require a lens of focal length f = 1 cm which is not too practical for an evaporator. Thus, the usefulness of the Q-switched mode has been eliminated.

This model leads to the suggestion that the target area should be smaller than or equal to the focused laser beam in order that complete vaporization takes place. Figure 5 illustrates two possible sample configurations. Measurements have been made with these configurations. With such targets, the ability to aim the laser beam accurately is very

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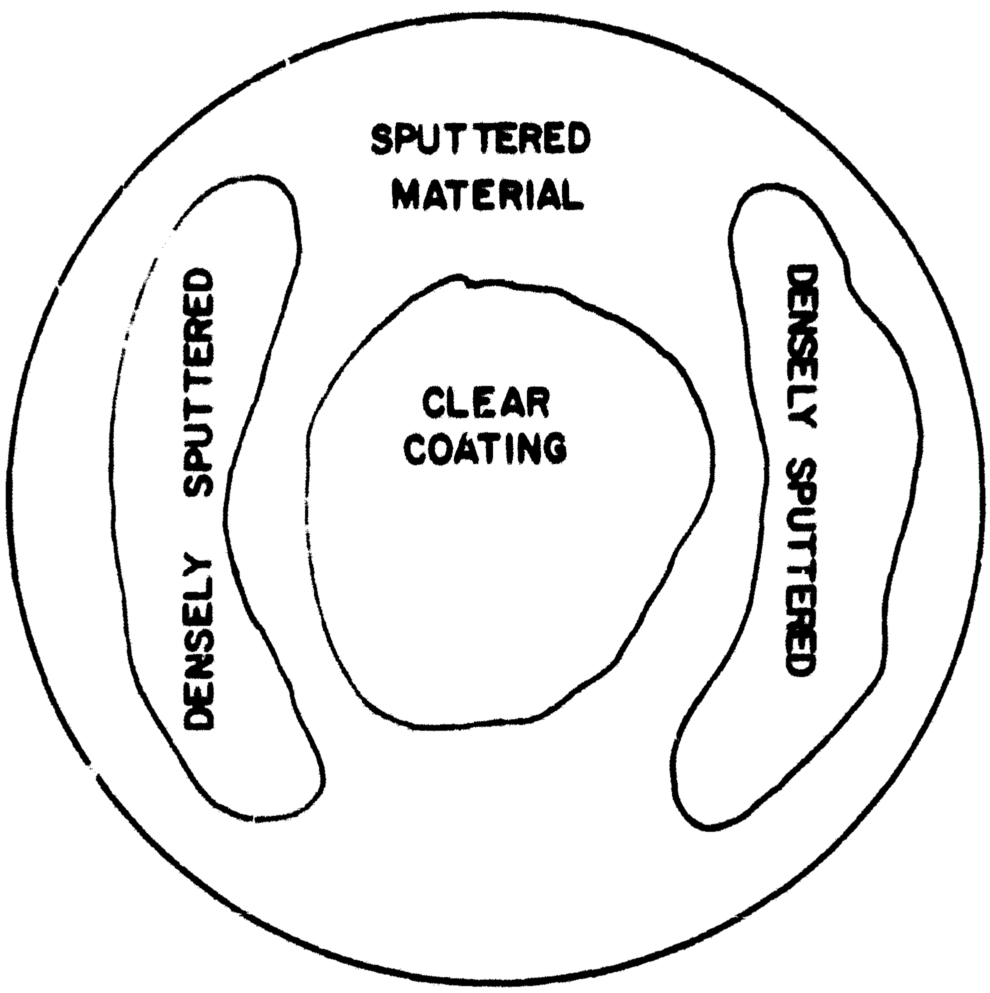

Figure 2. Diagram of the distribution of evaporated material on a hemispherical surface.

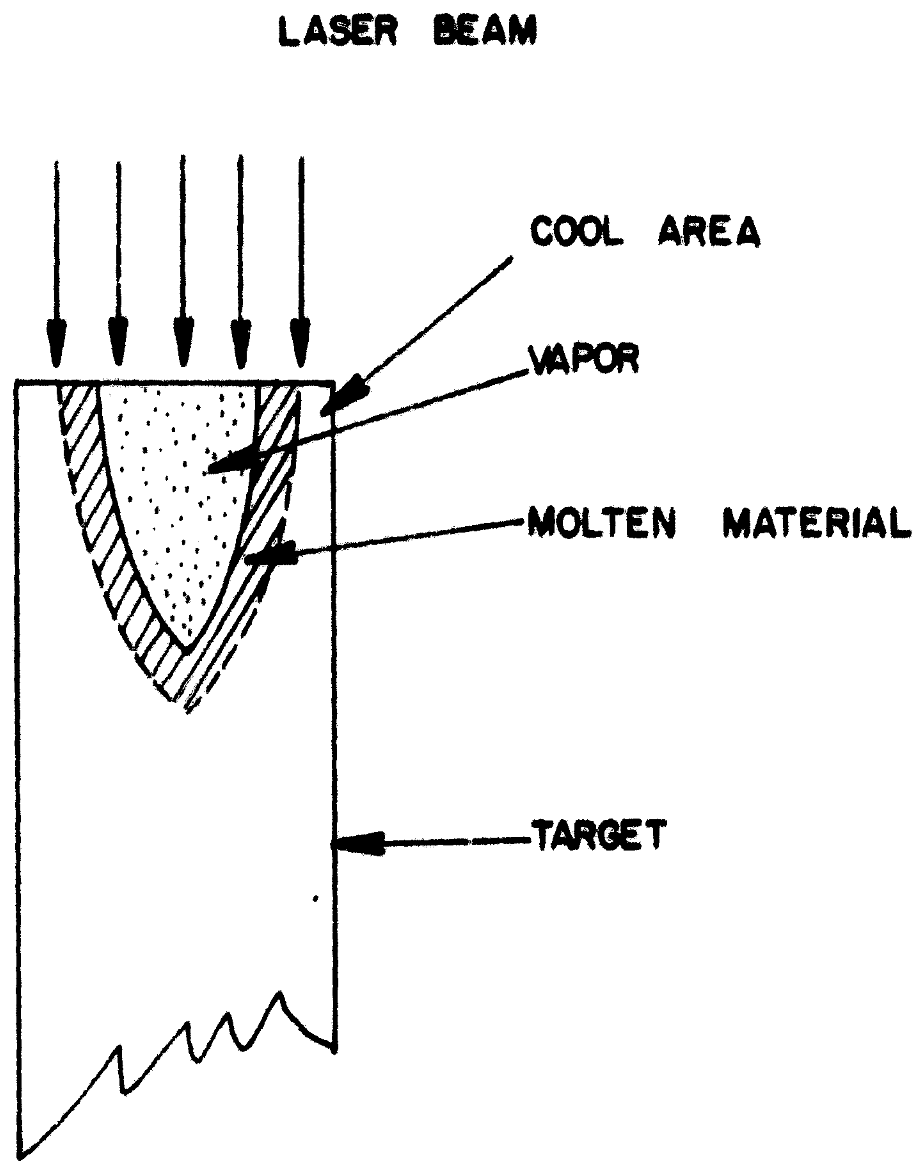

Figure 3. Suggested condition of solid, molten, and vaporized material on impact by a laser pulse.

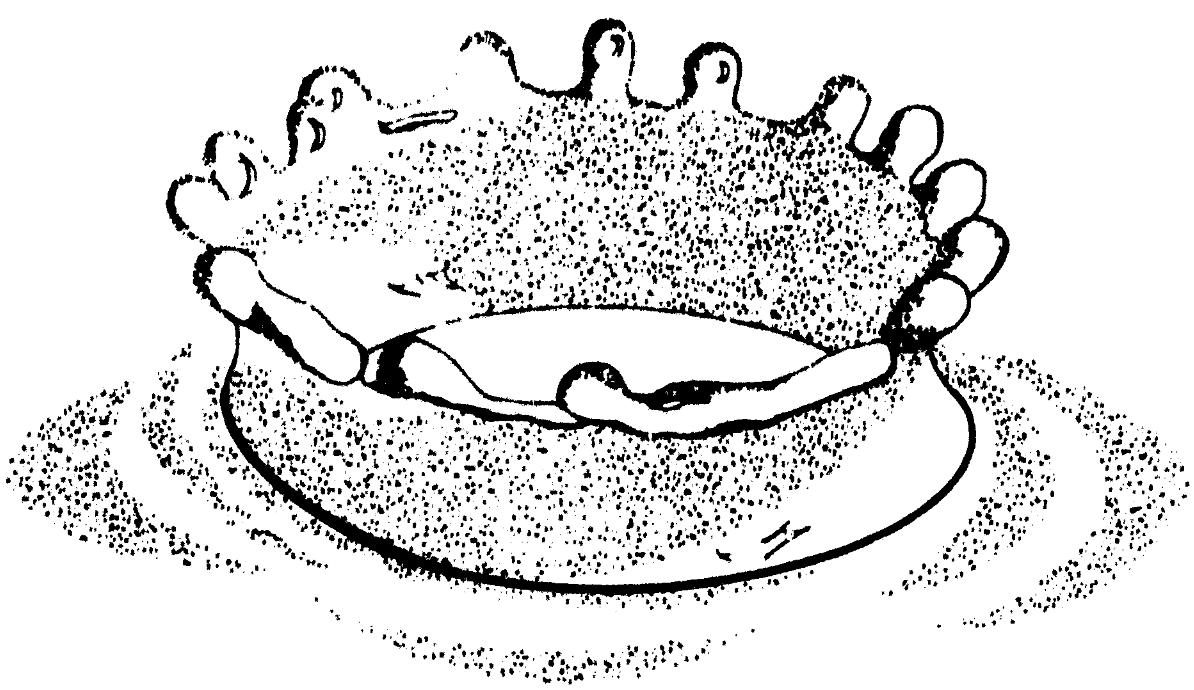

Figure 4. A shallow pool splash of a milk drop.

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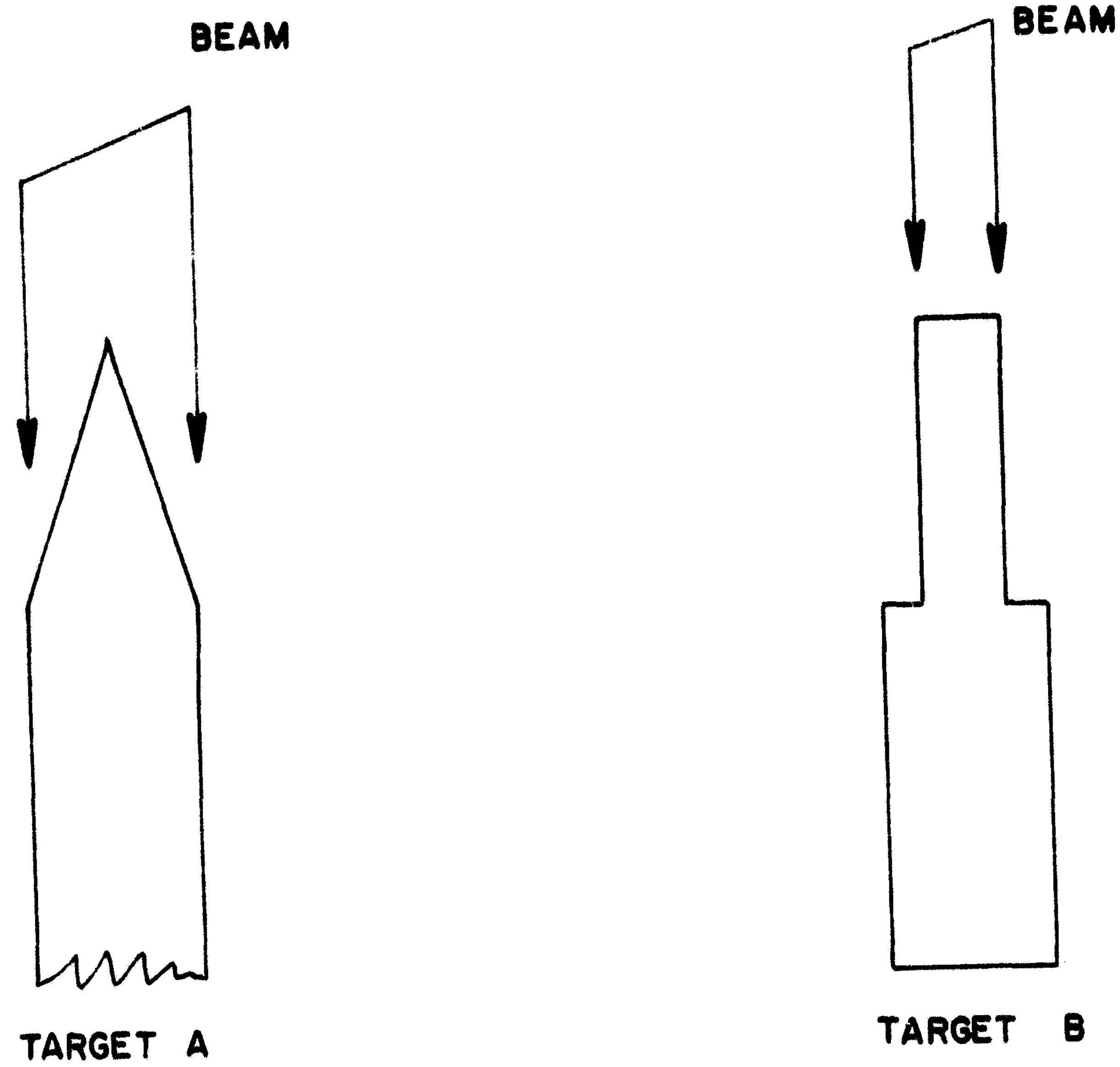

Figure 5. Targets used to study the ratio of sputtered-to-evaporated material.

important. It was found to be impossible to hit such a sharp target with the present arrangement since the curvature of the liter flasks was not uniform. Consequenly, a 1 inch hole was made in the flasks and a flat pyrex window was sealed in place. This procedure made a big difference in the ability to aim the laser beam accurately. Results with the sharpened point (target A) were disappointing. On reconsidering this design, it is obvious that a beam striking target A, as shown in Figure 5, would have a smaller flux density impinging upon unit area of the metal. However, using target B, the sputtering was considerably reduced. An aluminum cylinder 1 mm in diameter and 5 mm in length was used as shown in Figure 5. The focused laser beam had a diameter of 1 mm. Although the aim of the laser beam was not quite perfect, one shot nearly eliminated the entire 1-mm diameter cylinder. The disadvantage of such targets is the problem of making direct hits. However, if the aiming technique can be improved, then it would appear that more power density in the focused beam would be desirable since this should produce a higher ratio of vapor to liquid phase. The power density can be increased by using a higher input power or by sharper focusing of the laser beam. As described in the first section of this report, the diameter $\underline{d}$ of a focused laser beam is given approximately by

$$d = 2f\theta$$

where f is the focal length of the lens and θ is the angular divergence of the beam. The results reported herein used a lens of focal length f = 100 mm. Thus, $\underline{d}$ should be equal to 1 mm. This diameter, ±10 percent, was actually achieved in practice. Owing to the dimensions of the liter flasks, it is not possible to use lenses of extremely short focal lengths.

Other experiments were performed to reduce "sputtering" such as the use of a target made from tungsten foil one thousands of an inch thick folded in a zig-zag manner. It was hoped that each spike within a laser pulse would vaporize the thin foil. With this arrangement the laser beam punched a hole through all the layers present in the target (approximately twelve layers) but produced a negligible amount of vapor. A more densely packed zig-zag target produced the same result.

An auxiliary experiment was designed to test the effect of an electric field on the laser evaporated material. The experimental set up is shown in Figure 6. Various tests were made first with +5000 V, then with -5000 V. It was hoped that a marked effect on the ratio of vapor to "sputtered" material would be observed. Sometimes a positive voltage improved this ratio at other times negative voltages appeared more suitable. However, the density of a single pulsed evaporated coating on the slide always appeared to be about the same suggesting that if the vapor were charged the field configuration and voltage used was insufficient to affect the vapor. Whether the heavy sputtered material was affected by the field is difficult to say but the results showed that if there was an effect it was random.

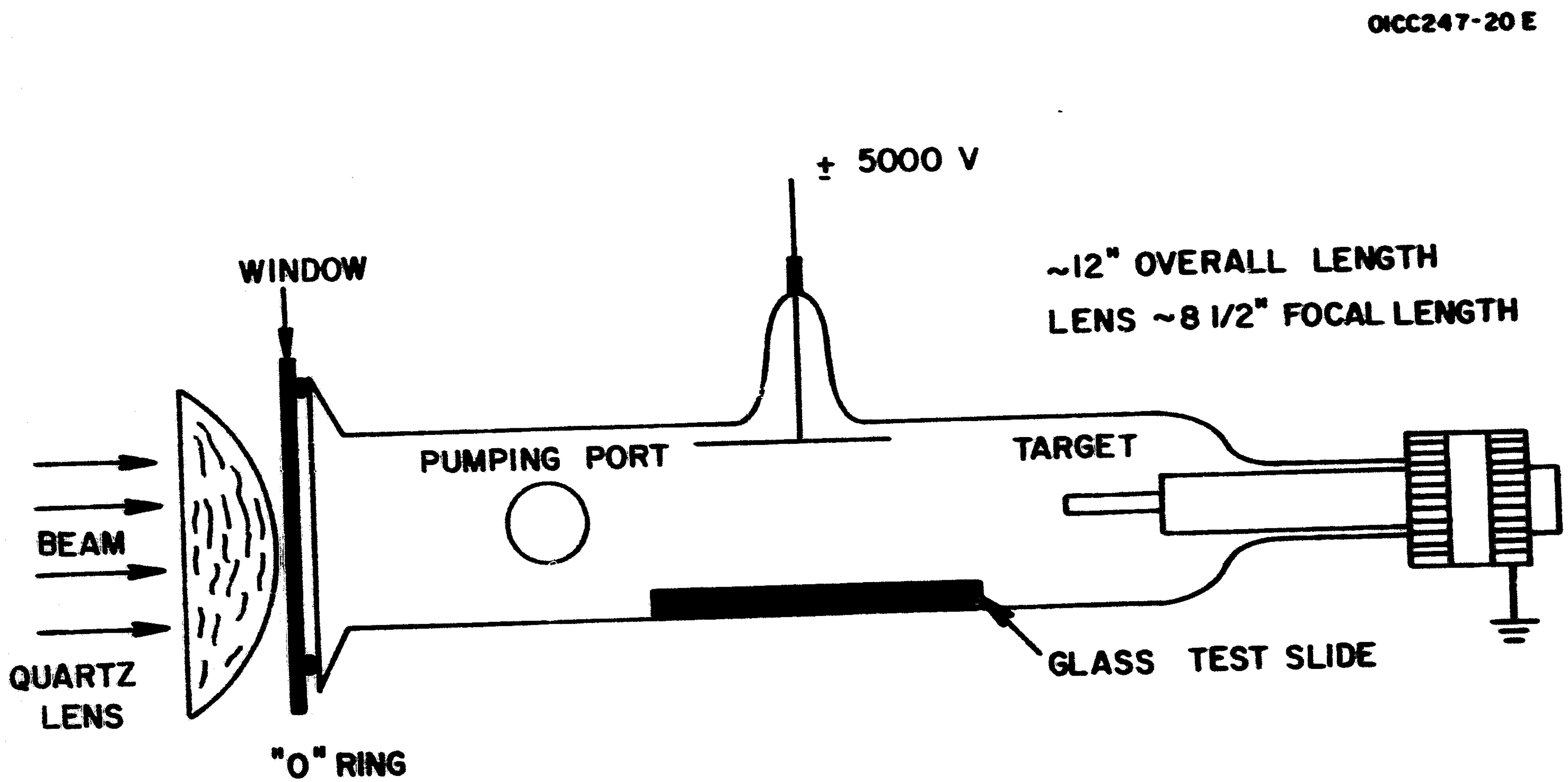

Figure 6. Evaporation system to study the effects of an applied electric field.

Another experiment used powdered tungsten as a target. The powder was placed in a boat and inclined at an angle of about 30° to the horizontal. No evaporation was noticeable. Each time a laser pulse hit the metallic powder it merely spilled the powder from the boat. This is further evidence that the laser pulse acts as a projectile when impinging on a metal.

The final experiment utilized a target made of compressed tungsten powder. The powder did not fuse but was sufficiently compressed to form a solid (but fragile) target. Evaporation was excellent and sputtering was greatly reduced. Compressed powdered molybdenum samples were supplied by the Goddard Space Flight Center. Like tungsten these samples gave excellent results.

Both tungsten and molybdenum films were evaporated using one, two and three laser burst of radiation. The mirrors produced by the three shots were estimated to have film thicknesses of about 500Å. Similar films were deposited on thin plastic scintillators (type Pilot B). By measuring the fluorescence produced first by an uncoated scintillator, then by a coated one, a measure proportional to the transmittance of the thin film was obtained in the vacuum uv spectral region. The results showed that both tungsten and molybdenum films 500Å thick were quite opaque to vacuum ultraviolet radiation above 600Å in wavelength. Molybdenum starts to transmit weakly around 530Å while the tungsten transmittance onset was about 500Å in wavelength. Thus meaningful reflectance measurements were feasible.

No reflectance data was taken for aluminum because of the excess amount of "sputtering" on the substrates. The reflectance of molybdenum and tungsten are shown in Figures 7 and 8. At wavelengths shorter than 600Å the reflectance decreases as is characteristic of most metals. However the reflectances are lower than that of published data on iridium or platinum especially at the short wavelength region. The reflectance data reported here can be expected to be lower than is characteristic of the metal since scattering of the radiation due to surface roughness is inevitable. Both mirrors exhibited some particulate material on their surface. However, the surface smoothness of the substrates is particularly important to prevent scattering of the shorter wavelengths. This has been shown to be the case theoretically by Bennett and his colleague (Ref. 1 and 2) and in practice by the Harvard College Observatory Group (Ref. 3). The glass substrates used for the data shown in Figures 7 and 8 were not optical flats nor were they treated for surface smoothness.

Good quality mirrors can be produced by a pulsed ruby laser and be used for research studies. The amount of "sputtered" material still present, although small, precludes the use of the mirrors in an optical system where the amount of scattered radiation must be kept to a minimum. The use of compressed powdered targets appears to be desirable when a pulsed ruby

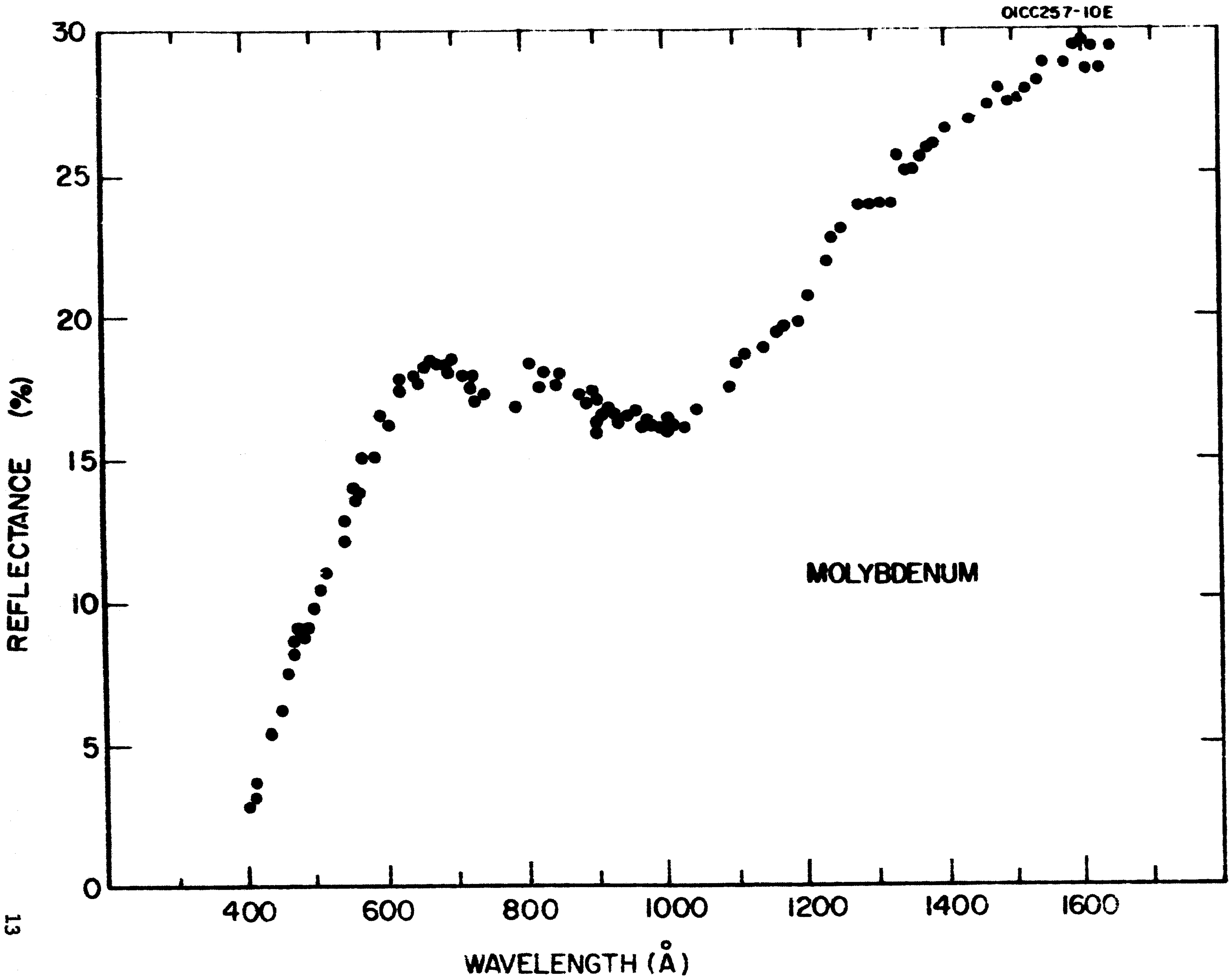

Figure 7. Reflectance of molybdenum measured near normal incidence

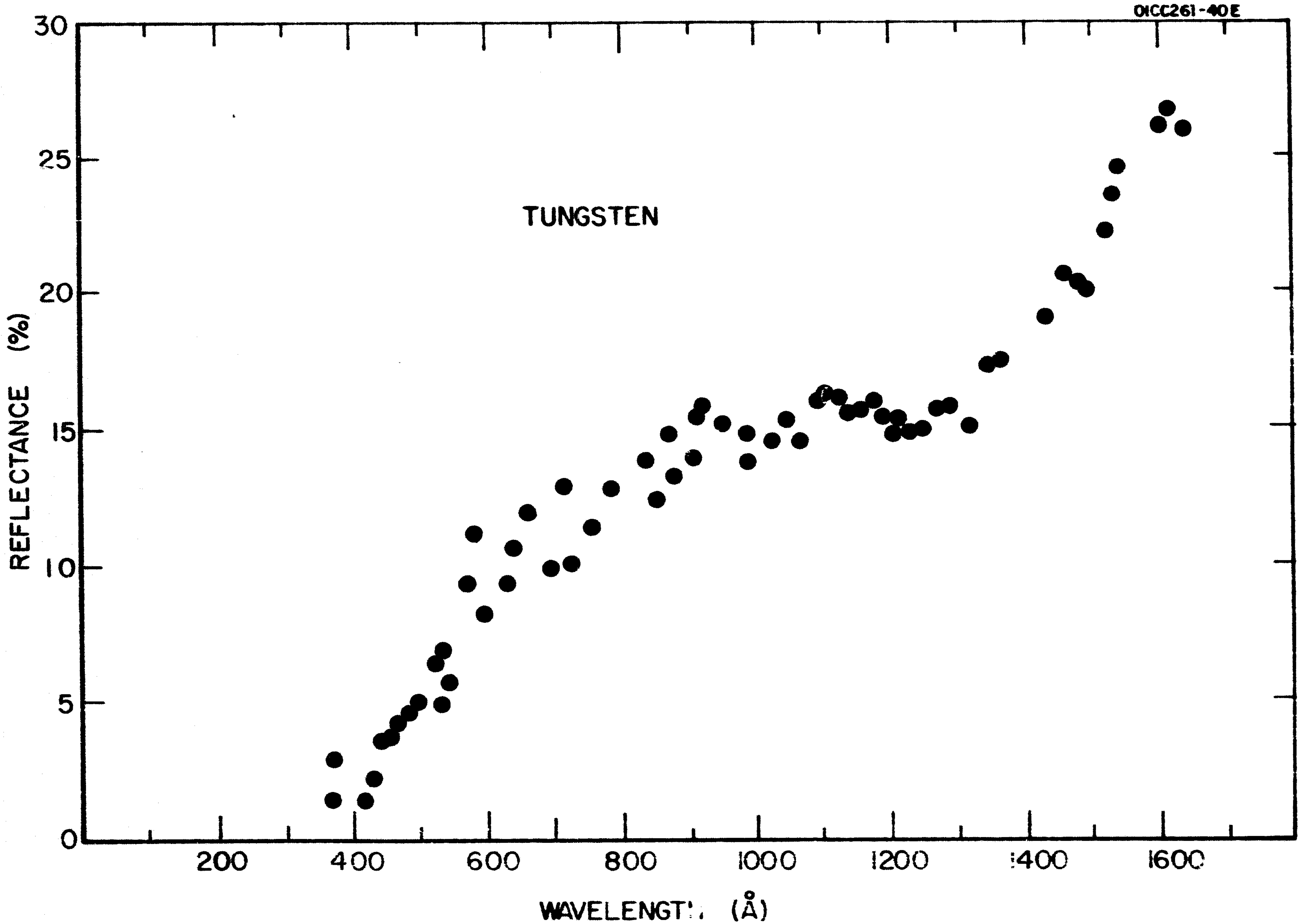

Figure 8. Reflectance of tungsten measured near normal incidence.

laser is used. Probably the use of a continuous wave laser of sufficient power such as the CO_2 laser will overcome the remaining problems of "sputtering." The technique of using a laser beam to evaporate materials has great potential in producing mirrors of the highest quality and purity, especially for use in ultra high vacuum systems.

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