# THE PRODUCTION AND DETECTION OF A MOLECULAR BEAM

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Robert Belson Colten

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This is to certify that the

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# THE PRODUCTION AND DETECTION OF A MOLECULAR BEAM

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L. B. Colton

# INTRODUCTION

The production and detection of a molecular beam, though it is only the groundwork for more elaborate experiments, in itself demonstrates an important physical principle, the straight-line motion of the molecules in a gas.

Experiments with molecular beams have contributed considerably to our understanding of molecular and atomic properties by offering a means of studying the effect of physical forces upon individual atoms or molecules. Among the important fundamental theoretical postulates which have been investigated by this method we might note the following: demonstration of the straight-line motion of gas molecules (as in this experiment), verification of the Maxwellian distribution of velocities of gas molecules, measurement of the mean free path, direct demonstration of space quantization, and proof of the wave property of material particles as postulated by de Broglie (with his relation $\lambda = h/mv$). Molecular beam experiments have also made possible the measurement of the following molecular and atomic properties: electric and magnetic moments, molecular collision cross-sections, and nuclear spins

and moments.

Such an imposing list of accomplishments could not have resulted but for the great patience and experimental skill of the men who have developed the molecular beam technique. However, the underlying principle involved is quite simple. If molecules, moving with thermal velocities, are introduced into an evacuated vessel they will travel in a straight line until they collide with another molecule or the walls of the vessel. If the pressure is low enough so that the mean free path is longer than the vessel, a beam is produced by placing another aperture in line with that through which the molecules entered (see Fig. 1). When a beam material which is condensable at room temperatures is used, a glass target can serve as a detector (for most metals).

In this experiment a beam of lead molecules was produced. A new oven design was tried which showed considerable promise. An oil diffusion pump was employed to evacuate the system with a Cenco Megavac furnishing the fore-vacuum. Pressure was measured with Pirani and McLeod gauges and the oven temperature was determined by measuring the resistance of the tungsten heating coil. The beam was detected with a condensation target, a clean glass plate upon which the molecules condensed (see Fig. 2). The project was undertaken in order to build and test a setup which might be used for various types of molecular beam experiments.

## DISCUSSION

A molecular beam consists of neutral particles moving with thermal velocities (about $10^4$ to $10^5$ cm./sec.) in a beam defined by a collimating system. Fraser (2) uses the terms "unidirectional" and "collision-free" to describe such a beam; both very closely approximating the facts. It must be clear that if neutral molecules can be made to travel in a well-defined path, the effect upon them of an inhomogeneous magnetic field, for example (Stern Gerlach experiment), can be observed. In one experiment a velocity selector was used to get a monochromatic beam which was reflected from a crystal to demonstrate that particles behave as waves with the wavelength which de Broglie had predicted. A great many different experiments have been performed with this method but the possibilities are far from exhausted.

Dunoyer's pioneering experiments in 1911 pointed to a whole series of investigations when he showed that a beam of neutral particles could be produced (6). O. Stern and his collaborators began in 1919 to employ and develop this tool with remarkable success. The literature contains reports from numerous investigators concerning the theory and results of molecular beam

experiments and the development of the various components thereof (see bibliography).

Even the simplest molecular beam experiment requires the following equipment: (1) a vacuum chamber with fast pumps to evacuate it and keep the pressure down, (2) a source of molecules, (3) a collimating system to define the beam, (4) some method of detecting the beam. It is also advisable to have some means of measuring the pressure in the system and of determining the oven temperature. Depending upon the type of experiment being performed, it may also be necessary to provide a magnetic field, electric field, crystal lattice, velocity selector, scattering chamber, and other pieces of equipment.

In most experiments the results are directly dependent upon the length of the beam. For example, when the beam is being deflected by some means, the effect is amplified by lengthening the distance from source to detector. There are two main limitations on the length of the beam: (1) the pressure of the system, (2) the sensitivity of the detector.

The pressure must be low enough so that the mean free path will be considerably longer than the distance traveled by the beam (at least twice as long), otherwise scattering due to collision with residual gas molecules would spoil the definition. Thus for a

setup thirty centimeters long the pressure should be lower than $10^{-4}$ mm. of Hg, which corresponds to a mean free path of about 65 cm. It must be borne in mind that the difficulties of obtaining such a low pressure are multiplied by the fact that the source is generally producing some extraneous molecules (similar and foreign) which must be removed.

Detector sensitivity becomes more important as the beam is lengthened because the beam intensity varies inversely as the square of the distance. There is a definite limit to the number of molecules which can leave the source in unit time through a given size aperture. Any attempt to increase the beam strength by raising the pressure in the source results in a jet or hydrodynamic flow when the mean free path in the source approaches the dimension of the aperture (7 and 8). The sensitivity of the detector, then, might become the limiting factor in the length of the beam and therefore in the effectiveness of the experiment.

## DESCRIPTION OF APPARATUS

The vacuum envelope consisted of a glass cylinder 30 cm. long with an outside diameter of 10 cm. and an inside diameter of 9 cm. which was waxed into annular groves in the brass end plates. This part of the apparatus was mounted in a vertical position with the

oven chamber soldered to the lower plate. Provision was made for pumping from both ends but only one line was used. Pirani and McLeod gauges were connected to the top and the pump line to the bottom (see Fig. 2).

The oven chamber consisted of two concentric brass cylinders 16 cm. long with water flowing between for cooling. The inside diameter was 3.75 cm. with a 5 cm. outside diameter. A hole 1 cm. in diameter was drilled into the vacuum envelope from the oven chamber through the 1.3 cm. thick end plate.

For an oven, a rock material called "Lava A" was selected which could be machined and then fired to make a good strong heat resistant ceramic.* Lead to be melted for a beam material was placed in a well in the center. Tungsten wire (.020 inch) was threaded through lengthwise holes around the center well to heat the oven (Fig. 3). Three threaded rods were used for mounting the oven on the bottom plate of the chamber so that it could be adjusted to any desired position in the chamber.

The bottom plate was bolted to the oven chamber and sealed with a rubber gasket in between. One heater connection was brought through the bottom plate by means of a copper rod threaded through a

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*Obtained from American Lava Corp., Chattanooga 5, Tenn.

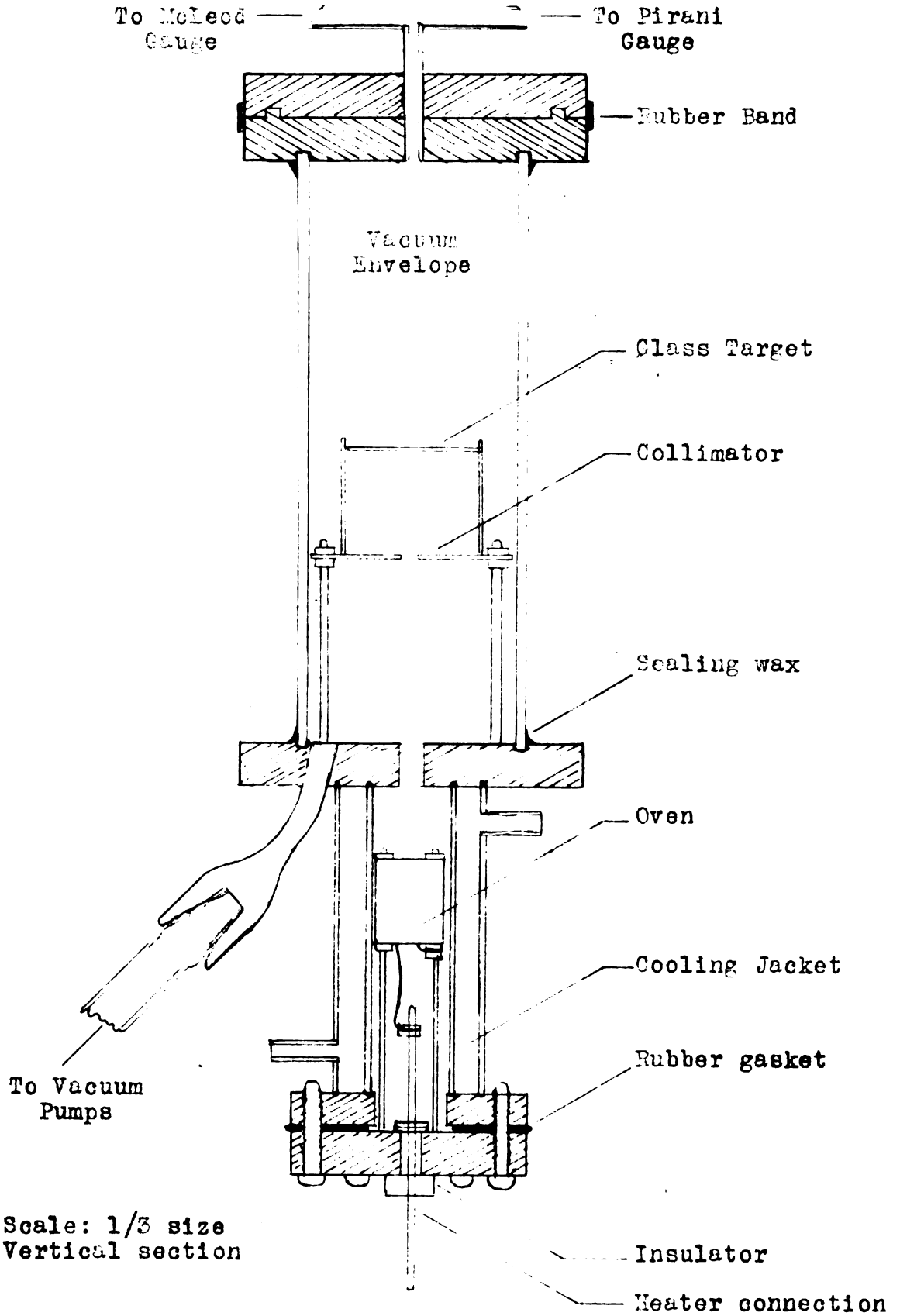
To McLeod Gauge
To Pirani Gauge
Rubber Band
Vacuum Envelope
Glass Target
Collimator
Sealing wax
Oven
Cooling Jacket
Rubber gasket
To Vacuum Pumps
Scale: 1/3 size
Vertical section
Insulator
Heater connection

FIGURE 2

fiber insulator sealed into the brass bottom plate with Glyptal. The other connection was made to the plate.

An all metal single jet oil diffusion pump with a self-contained electric heating element pumped against a fore-vacuum produced by a Cenco Megavac pump. Though it is not always considered necessary, a trap was inserted between the diffusion pump and the envelope. Another trap was used in the line to the McLeod gauge. Both traps were cooled with a mixture of dry ice and acetone.

The oven heater element was connected to a double pole double throw switch so that it could be alternately connected to a "Variac" autotransformer or to a Wheatstone bridge as desired. By switching over from the "Variac" to the Wheatstone bridge and getting a quick reading of the resistance, the temperature of the oven could be estimated.

For one experiment a steel plate 0.3 mm. thick with a 1/32 inch hole was used as an oven aperture. Another metal plate with a 1/4 inch hole was placed 3 cm. from the target. The target was a clean glass plate 15 cm. from the oven aperture (Fig. 2). The design was such that the tungsten element heated the steel plate to a temperature slightly higher than the rest of the oven to keep the hole from becoming plugged with condensed metal. It was assumed that molecules

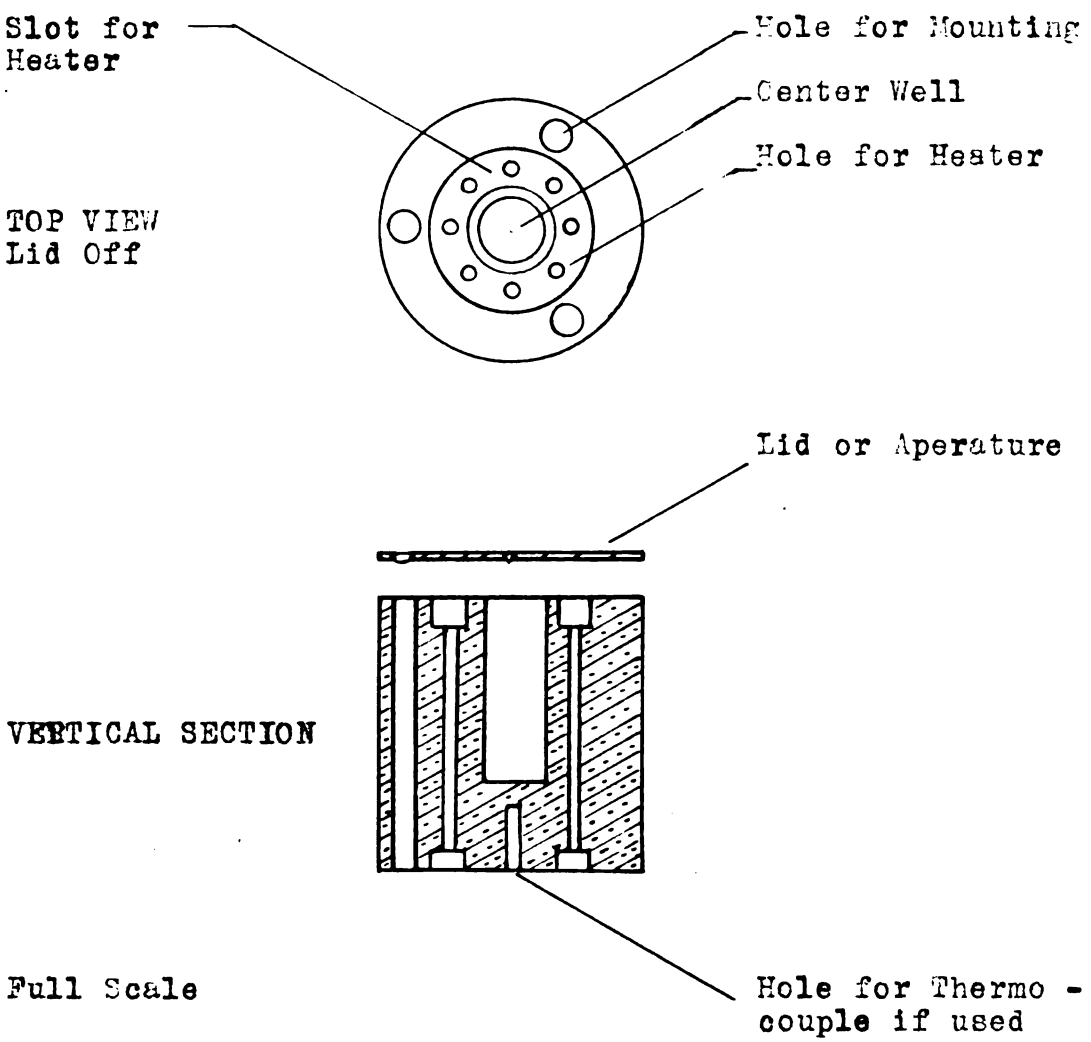
Slot for Heater
Hole for Mounting
Center Well
Hole for Heater
TOP VIEW
Lid Off
Lid or Aperature
VERTICAL SECTION
Full Scale
Hole for Thermo - couple if used

FIGURE 3

which struck the walls of the envelope, the collimator plate, etc. would condense and not interfere with the beam.

A seal at the top end of the vacuum envelope was made with a rubber band which was stretched over the crack between two closely fitting plates and painted with liquid polystyrine. The brass plates, one waxed to the glass and the other connected to the vacuum gauges, were made with an annular tongue and groove to prevent sidewise motion. A good vacuum seal was thus provided but ready access to the interior was available.

## OPERATION AND RESULTS

With the oven loaded and the system sealed, the forevac pump was started. Dry ice and acetone was placed in the traps, cooling water to the diffusion pump was turned on, and the diffusion pump heater was energized. When the diffusion pump took hold, the pressure would fall rapidly to a value too low to be read on the Pirani gauge but would level off somewhere above $10^{-4}$ mm. Hg. Considerable pumping and sparking with a Tesla coil (to outgas) finally brought the pressure down below $10^{-4}$. This was not accomplished, of course, until a number of attempts had been made

and several leaks located. It was observed that the voltage on the diffusion pump heater was quite critical, or, in other words the oil temperature must be just right for most efficient operation.

Cooling water was started to the oven chamber and current was turned to its heating coil. Even with a small current flowing, pressure would rise rapidly and level off at some value (with 5 amperes, pressure rose to 0.1 mm. Hg) but the pressure would go down slowly as degassing took place. It was found that by intermittently turning the oven heater on and off (on 15 minutes, off 1 minute) the current could be raised to the desired value by steps, and the pressure brought down faster. About 150 watts was required to operate the oven at the desired temperature.

It took 24 hours from the time the pumps were started until an indication was noticed on the target. Eight hours later, a well defined dark spot had developed. During the effective part of this experiment the oven was held at about 550° C. and the pressure in the vacuum envelope was below $10^{-4}$ mm. Hg.

The temperature of the oven was computed from the change in resistance of the heating element, using the common formula:

$$t = \frac{r - r_o}{r_o \alpha} \qquad \text{Equation 1}$$

where: t is the temperature rise in degrees C.,

$r_o$, the original resistance in ohms,

r, the final resistance in ohms, and

$\alpha$, the coefficient of resistivity in ohms/degree. A value for $\alpha$ for temperatures around $500^o$ was given in the handbook (18) as .0057 ohms/degree. With the resistance of the heater rising from .161 ohms to .635 ohms, the temperature was figured to have been about $550^o$ C.

The spot on the target had a dark, bluish central portion about 7 mm. in diameter, surrounded by colored interference rings when viewed in a white light. The rings were caused by a thin film effect indicating that a certain amount of diffuse scattering had occurred in the beam. The deposit was thin enough to be translucent to normal light rays, showed gray metallic surface in shaded light, but was quite visible from almost any angle.

There are two possible causes for the scattering observed above; (1) the pressure in the vacuum envelope may have become too high at some time during the experiment so that collisions took place in the beam path, or (2) the pressure in the oven might have been high enough so that the mean free path in the oven approached the order of magnitude of the aperture, in which case hydrodynamic flow would take place with the resultant "cloud" formation in front of the aperture.

During one period the temperature of the oven was raised to 700° C. to get a check with an optical pyrometer.

The mean free path in the oven at 550° C. was determined to see how it compared with the size of the aperture. A good approximation for the value of the mean free path is given by the expression (17);

$$L = \frac{1}{\pi N d^2} \qquad \text{Equation 2}$$

where: L is the mean free path in cm.,

d, the diameter of a molecule (or atom, since lead is monatomic),

N, the number of molecules per $cm^3$.

A value for the diameter of a neutral lead atom was found in the handbook (18) to be $3.48 \times 10^{-8}$ cm. To obtain a value for the number of molecules it was first necessary to determine the vapor pressure of lead at 550° C. This was computed from the formula (18):

$$\text{Log}_{10} P = \frac{-0.05223\ a}{T} + b \qquad \text{Equation 3}$$

where: P is the pressure in mm. Hg,

T, the absolute temperature,

a, 188,500 for lead,

b, 7.827 for lead.

Equation 2 gives a value for the vapor pressure equal to $6.5 \times 10^{-4}$ mm. Hg.

Having determined the pressure in the oven we can find the number of molecules from the gas laws, knowing that at the same temperature and pressure all gases contain the same number of molecules in a given volume. Thus

$$N = M \cdot \frac{P_n}{P} \cdot \frac{T_m}{T} \qquad \text{Equation 4}$$

where: N is the number of molecules per $cm^3$ at $550^\circ$ C. and $6.5 \times 10^{-4}$ mm. Hg.

M is the number of molecules per $cm^3$ of an ideal gas at $0^\circ$ C. and 760 mm. Hg.

A value for M was derived from the fact that there are $6.02 \times 10^{23}$ molecules per mole of any substance and one gram molecular weight (mole) of an ideal gas occupies 22.4 liters. The $M = 2.71 \times 10^{19}$ molecules/$cm^3$.

$P_n$ and $P_m$ are the pressures in mm. of Hg corresponding to N and M.

$T_n$ and $T_m$ are the absolute temperatures corresponding to N and M. Equation 3 gives a value of $5.9 \times 10^{12}$ molecules/$cm^3$. for the number of molecules in the oven vapor.

If the above values are used in Equation 2, we find that the mean free path in the oven is about 45 cm., considerably larger than the size of the aperture which was about 0.08 cm. Thus we see that the oven pressure was not too high and it is quite unlikely

that any turbulence was produced at the aperture.

If we assume that we had all the necessary conditions for a beam during a period of 24 hours and that the oven held a constant temperature of 550° C., we should be able to compute the thickness of the deposit that would be formed under normal conditions. It is reasonable to say that practically every molecule which reached the target adhered to its surface.

The number of molecules, dN, which strike a surface element, dS, on the target is given by (3):

$$dN = \frac{5.83 \times 10^{-2}\ a\ p\ ds}{r^2\ (MT)^{\frac{1}{2}}} \text{ moles/sec.} \quad \text{Equation 5}$$

where: a is the oven aperature in $cm^2$. (diameter 0.8 mm.),

p, the oven pressure in mm. Hg equal to the vapor pressure of lead at 550° C. (about $6.5 \times 10^{-4}$ mm. Hg),

r, the distance from oven to target (15 cm.),

M, the molecular weight of beam material (207),

T, the absolute temperature of the oven (823° K.).

From this we find that $7.5 \times 10^{-13}$ moles/sec. reach the target, which corresponds to $1.6 \times 10^{-10}$ grams/sec. At this rate we should get a deposit 300 Angstroms thick, or, roughly, 100 atoms thick in twenty-four hours. The deposit actually formed appeared to be somewhat less

than 300 Å thick but of that order of magnitude.

It is apparent from the above figures that quantitative measurement of such results would be quite difficult. Use of micro-balance has met with little success. Optical methods are hampered by the fact that the deposits do not form in a uniformly thick layer (10 and 11).

The oven caused some trouble, burning out first a nichrome and then a tungsten heater. But the final heater wire showed no deterioration except for the usual brittleness after heating, though it was exactly like the previous tungsten one which had burned out. It is possible that in the firing process some water had been left in the ceramic material which reacted with the tungsten.

On one occasion, when the oven was removed after a forced shut-down, the aperture was found plugged with lead. This may have occurred after the heat was off and thus have no significance. If the hole became plugged while the heat was on, which does not seem too likely, it would indicate that the front plate was not sufficiently heated. In designing an oven it is essential to be sure that the aperture will be the hottest part.

Provision could have been made for pumping directly from the oven chamber as well as from the vacuum envelope. The advantage of such an arrangement must

be weighed against the increased complication of an additional diffusion pump.

## CONCLUSION

We have produced and detected a molecular beam and demonstrated that the results were approximately what would have been expected from theoretical considerations.

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