MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1965-A
CLUSTER BEAM STUDIES

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Cluster beams offer a means of depositing high-quality thin films at low substrate temperature for microelectronics fabrication. The advantage of cluster beam depositions is the ability to optimize the energy of the impacting particles, either directly in clustered vapors of nonvolatile materials or indirectly by bombarding the film during deposition with clusters of inert gases. When a cluster beam is ionized and accelerated through several thousand volts, clusters that contain 1000 or more atoms strike the surface with several electron volt energy per atom. The suprathermal energy of the depositing atoms is thought to produce unique thin films (either in quality, or in the ability to be deposited at all). This report describes the general effort on cluster beam formation methods, on cluster ionization by electron bombardment in a gridded ionization cell, on electrostatic mass-separation, and on electrostatic acceleration to a predetermined velocity. Detailed results are given on the improvements in performance of ionization cells for cluster beams of nonvolatile and gaseous materials.
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INTRODUCTION

This annual report presents the results obtained under the Cluster Beam Studies Program being performed for the Air Force by Hughes Research Laboratories under Contract F49620-85C-0125 DEF. The work under this Air Force Program is being conducted concurrently and coordinated with broader-based cluster studies being performed at Hughes under Hughes IR&D and Navy Contract N-000014-86-C-0705. Some of the relevant results from these other programs are also described and discussed in this report to place the results obtained under the Air Force Program in an overall perspective.

Work on ion cluster beams began at Hughes in 1983 with an investigation of a low-temperature deposition process for microcircuit fabrication, using ion cluster beams as reported by T. Takagi and members of his laboratory at the University of Kyoto in Japan. According to Takagi, ionized clusters are effective vehicles for thin-film deposition because large atomic fluxes of pure materials can be directed to the deposition substrate with controlled kinetic energy. Energies of a few electron volts per deposited atom are thought to be essential to growth of high-quality films at low substrate temperatures. The exact mechanism by which energetic particles enhance high-quality film growth is not fully understood. It is plausible that the depositing atoms are rearranged into more uniform and compact arrays (crystalline or polycrystalline) because of the energy imparted to the film by the incident flux of energetic ionized clusters, even though only a small percentage of the total incident flux is comprised of clusters. If this supposition is valid, it should be possible also to improve thin film growth indirectly by using energetic clusters of an inert gas to bombard the substrate while the film is formed by vapor deposition. This alternate deposition concept (invented at Hughes Research Laboratories) has been called cluster-assisted deposition. At present, both the direct ion cluster-deposition approach,
described by Takagi, and the cluster-assisted deposition approach are being studied at Hughes.

Whereas most of the work reported in Japan places emphasis on deposition of films or growth of materials using ion cluster beams, the work at Hughes has concentrated on defining criteria for formation of clusters and documentation of ion cluster-beam properties. Unfortunately, reports on work in Japan contain very little description of measurement or control of the conditions that prevail during the deposition processes. In fact, the average cluster size and the composition of the deposition flux with regard to clusters or ions is not reported and, at least in some instances, was not known. On the other hand, the approach at Hughes places priority on developing an understanding of ion cluster-beam properties before attempting an exhaustive program of film deposition or unique material preparation, even though the ultimate objective is to develop a high quality film deposition process for submicrometer electronics applications.

At the start of the Air Force Cluster Beam Studies Program, considerable progress had already been made towards analysis, design, and fabrication of experimental apparatus to investigate cluster beam formation from vapors of both volatile and nonvolatile materials. Enough experience had been gained in nucleation, ionization, and beam transport and deposition to identify the technical problems in each of these areas. Based upon these assessments, the objective of the first year of work under the Air Force Program was to improve the performance of the ionization cell for cluster beams of both nonvolatile and volatile materials.
This report has been organized so that the first sections summarize progress under all the cluster studies at Hughes Research Laboratories (funded by Hughes IR&D and ONR Contract N-000014-86-C-0705) and the final sections describe the results obtained under the Air Force Program.

ION CLUSTER-BEAM STUDIES AT HUGHES RESEARCH LABORATORIES

Ion cluster-beam studies at Hughes Research Laboratories can be divided into two categories according to whether the clusters are formed from volatile or nonvolatile materials. For volatile materials, or gases, the studies are directed towards cluster-assisted deposition using inert-gas clusters to impart energy to the surface where deposition is being performed. For nonvolatile materials, the current focus is on achieving precise control of cluster formation and cluster ionization in gold vapor for deposition on gallium-arsenide substrates.

Cluster Beams in Volatile Materials

Formation of clusters in volatile materials is achieved by isentropic expansion of gas at relatively high pressure through a supersonic nozzle into vacuum, as shown in Figure 1. A parametric study of this type of cluster formation has been reported by Hagena and Obert for the inert gases. All of the gas atoms and clusters in the central cone of the flow field are accelerated to the same velocity, and this velocity is a function only of the gas species and the nozzle temperature. The nozzle throat diameter, the nozzle shape, and the reservoir pressure determine the gas flow rate and vacuum pumping requirement for achieving isentropic expansion in the nozzle. For a given nozzle, the average cluster size is a function of the reservoir pressure.

At Hughes, the study of volatile clusters began with the construction of a gas valve and nozzle configuration like that
Figure 1. Illustration of cluster formation in gas expanded through a supersonic nozzle.
shown in Figure 2. Initial measurements were made to ensure that the cluster-size versus reservoir-pressure relationships, reported by Hagena and Obert for inert gases and nitrogen, could be reproduced in the experiments at Hughes. These measurements were made by ionizing the gas clusters and measuring the ion current as a function of the retarding potential applied to the Faraday cup grid using the experimental configuration shown schematically in Figure 2. An idealized current-voltage characteristic representative of these measurements is shown in Figure 3. In these experiments, all ions are accelerated by a voltage corresponding to the anode voltage of the ionizing cell, and the average cluster size, \( N_a \), (in number of atoms per cluster) is defined by

\[
N_a = \frac{2e (V_2 - V_1)}{m_0 v_s^2}
\]

(1)

where \( e \) is the electronic charge, \( V_1 \) is the anode voltage, \( V_2 \) is the voltage for which the collected ion current is one-half its maximum value, \( m_0 \) is the atomic mass of the gas species, and \( v_s \) is the limiting velocity of the gas (as determined by the nozzle expansion). For argon gas, Eq. (1) becomes

\[
N_a = 19.3 (V_2 - V_1)
\]

(2)

In addition to determination of the average cluster size, data, like that shown in Figure 3, can be reduced to obtain the distribution of cluster sizes. As mentioned previously, the use of ion clusters in microelectronic fabrication processes requires that atomic ions and small cluster ions be removed from the ion beam that is accelerated to high voltage and directed at the substrate being processed. In the cluster beam configuration shown in Figure 2, the grid that is labeled number 3 is referred to as the extraction grid of the ionization cell. This grid can
Figure 2. Ion cluster-beam experimental configuration.
$N_a = \frac{2e (V_1 - V_2)}{M_o V_s^2}$

$N_a =$ ATOMS/CLUSTER (AVERAGE)

$M_o =$ MASS OF GAS ATOM

$V_s =$ SUPersonic expansion speed

$V_2 =$ Voltage at which current is one-half of maximum value

Figure 3. Representative data obtained in experiment to determine average cluster size, $N_a$. 

13
be used to prevent the atomic ions and smaller cluster ions from leaving the ionization cell by means of electrostatic retardation. At the low-plasma densities that were produced in early experiments, the space potential in the ionization-cell plasma reached an equilibrium value that was slightly higher than the anode potential. Electrostatic mass separation was achieved simply by biasing the extraction grid at a voltage sufficiently more positive than the ionization-cell anode so that the extraction grid was more positive than the plasma space potential. For argon clusters, Eq. (2) can be used to estimate the cluster size required to overcome a bias voltage $V_2$ for operation of the ionization cell at a space potential, $V_1$. For example, if this potential difference is adjusted to be 20 V, then cluster ions will have to contain at least 400 atoms to overcome the extraction-grid bias and become part of the ion cluster beam. Figure 4 illustrates the representative data obtained and the cluster-size distributions produced in experiments to demonstrate this approach to mass selection. It is apparent that this approach assumes that all ions are generated at the same, or nearly the same, space potential. Any appreciable variation in the plasma space potential would have an adverse effect on this approach to mass separation and, while a few volt-variations are tolerable, variations of tens of volts cannot be accommodated. This subject will be addressed again in the discussion of ionization cells for cluster beams of nonvolatile materials.

Gas-flow distribution measurements document the atomic density as a function of distance from the nozzle throat. Pressure was measured in the gas-flow using a thin metal ribbon. The latter was stretched in front of a highly sensitive capacitance bridge that measures distance. This pressure sensor was mounted on an x-y stage as shown in Figure 5. Figure 6 represents the pressure distribution as a function of distance from the nozzle. Gas-valve operating parameters and nozzle-throat dimensions were varied until the valve could be operated
Figure 4. Measurements showing capability for achieving mass separation by biasing extraction grid relative to ionization-cell anode.
Figure 5. Experimental configuration for measurement of gas flow (pressure) distribution produced by nozzle.
Figure 6. Pressure distribution measured with conical nozzle and pressure probe.
at a 5 to 10% duty cycle with an acceptable increase in the background pressure (i.e., one that does not invalidate the conditions for isentropic expansion of gas in the nozzle).

Numerous experiments were performed to investigate ionized clusters generated in the basic configuration shown in Figure 2. Initially, these experiments had the primary objective of documenting cluster size as a function of reservoir pressure and gas species with different nozzle geometries. Little attention was paid to the ionizer efficiency or beam-formation characteristics. When evaluating cluster-assisted deposition, it was found that the original apparatus had limitations in the level of accelerating voltage that could be applied and in the ion current that could be produced by the ionizer. In the configuration shown in Figure 2, the entire ionizer assembly, comprised of the electrodes labeled 1, 2, and 3, is operated at high positive voltage (2000 to 6000 V is desired for ion clusters containing 2000 atoms). At gas-pulse repetition rates of 10 Hz, the gas pressure in the region between skimmer and ionizer rises to a value where Paschen breakdown can take place. This danger of breakdown limited not only the levels of accelerating voltage, but also the achievable rates of ion production, since an increase in the filament emission to increase ionization tended to lower the breakdown voltage. As will be seen below, the problem has been addressed under the Air Force program and considerable progress has been achieved.

Cluster Beams in Nonvolatile Materials

Work at Hughes on nonvolatile clusters began with the construction of a cluster facility for silver, patterned after the cluster sources described by Takagi. Initial film-deposition tests yielded unsatisfactory results, and a broad evaluation of the Takagi process was initiated. Measurements in the exhaust of the cluster-producing crucible showed that only about 1% of the
flux was comprised of clusters. Furthermore, the ionizer, in combination with an electrostatic accelerator, resulted in a beam with very uneven lateral distribution. Finally, the extracted ion beam contained many more atomic ions than cluster ions. Atomic ions must be considered detrimental to the deposition process, since they possess energies of thousands of electron volts in beams that have been accelerated to voltages where singly ionized-clusters possess only a few electron volts per atom. A few electron volts per atom is considered beneficial to the quality of film growth, whereas hundreds or thousands of electron volts are excessive, resulting in atomic displacements (point defects).

To overcome these and other deficiencies of the Takagi process, a better understanding of the basic cluster-beam process was sought. It was considered essential to address the following aspects of the process: cluster formation mechanisms, the cluster state, cluster ionization, cluster mass separation and acceleration, and cluster/surface impact processes. Significant progress has been made in several of these areas.\(^3\)

At the outset of this study, the possibility of large metal-vapor cluster generation by homogeneous nucleation in a nozzle exhaust, as postulated by Takagi, was viewed with considerable skepticism. While the generation of clusters in gases by this process is well-established, the pressures required in the cluster formation region are in the atmospheric range, whereas the pressure of the metal vapor in the nozzle apertures, where clusters are observed in the Takagi approach, can only be raised to a few Torr. Furthermore, cluster formation in gases depends critically on a high-nozzle expansion ratio, while cluster formation in metal vapors appears to be insensitive to nozzle shape. Sonic nozzles, with very limited expansion-cooling capabilities seem to be as effective as supersonic nozzles for cluster formation in metal vapors.

To better understand the differences between cluster formation in volatile gases and nonvolatile metal vapors, a
theoretical study of possible physical processes began. A first study object was cluster growth in a flowing vapor. A collisional model was adopted that was based on the random accretion of vapor atoms on the cluster and consideration of the removal of the heat of condensation through re-evaporation of some of the arriving atoms. The latter effect turned out to be a significant growth retardant, since many collisions are required for removing the latent heat associated with the condensation of a single atom. Because of this constraint, homogeneous nucleation in metal vapors is not expected to generate clusters larger than about 50 atoms under the conditions used by Takagi and also in the first experiments at Hughes.

A second possible growth mechanism analyzed was heterogeneous nucleation and growth (i.e., cluster formation on surfaces). The following process steps were considered essential to this mechanism: (1) formation of an adequate supply of critical embryos, (2) growth of embryos into full-sized clusters, and (3) ejection of these clusters into the surrounding vapor atmosphere. First of all, it was established that these growth steps can occur only with vapors of materials which do not wet the growth surface. 

Cu, Ag, Au, Zn, Cd, Hg, Ga, In, Tl, Ge, Sn, Pb, As, Sb, Bi, Se, and Te provide nonwetting combinations with such high-temperature crucible materials as graphite and boron nitride.

Theoretical expressions were derived for the rates associated with the three steps stated above, and these expressions were evaluated numerically for a few nonwetting material combinations, including Ag on graphite and Bi on graphite for which the atomic-attachment energies are known. In these cases, the three steps occur at a high rate and the predicted cluster-production rates agree with Takagi's observations. A peculiarity of the process for cluster generation on surfaces is that it proceeds only within a narrow range of temperatures. Only if the surface is a few degrees lower than that of the vapor-generating melt are
large clusters with hundreds to thousands of atoms produced. At present, crucible and nozzle configurations are being developed which take advantage of this property. The basic approach is to provide temperature gradients along the length of the crucible such that specific growth zones are obtained. A first test-crucible providing such a gradient has been built and initial test results appear to correspond to predictions of the heterogeneous nucleation process.

AIR FORCE CLUSTER BEAM STUDIES PROGRAM

The first year of the Air Force Cluster Beam Studies Program focused on development of improved ionization cells for cluster beams using both volatile and nonvolatile materials. Work on ionization cells under this program is coordinated with activities under the Hughes and Navy programs so that the hardware developed for formation of cluster beams can be used without duplication. As in the case of cluster formation, problems encountered in improving the ionization cell are different for volatile and nonvolatile materials. Consequently, the progress described here is also divided into ionization cells for volatile and nonvolatile materials.

Ionization Cells for Volatile Materials (Gases)

Work began under the Air Force program by continuing the evaluation of the initial ionization cell that was described earlier. Experiments were performed using argon gas with the objective of achieving the maximum ionization fraction without altering the cluster size distribution. The source was operated under conditions that produce average cluster sizes of 1500 to 2000 atoms. It was found, however, that the ionization cell had to be constructed with electrodes that have relatively high transparency to neutral gas flow, or the gas density would increase in the path of the cluster flow to the point where cluster/gas-atom collisions caused rapid decay of the clusters.
This was discovered experimentally when the highly transparent grid anode that encloses the cluster stream was replaced by a cylindrical anode that had a gridded opening only adjacent to the filament. While the latter geometry produced an increase in the ion current, it contained no appreciable fraction of large clusters.

The gas pressure (neutral atom density) in the ionization cell is relatively high, even with a relatively high-transparency ionization cell structure and, consequently, a dense, space-charge neutral plasma is always produced. Therefore, the space potential within the ionization cell is expected to be relatively uniform as required for mass separation and beam formation (discussed earlier). However, problems of a different nature arise from the presence of a high plasma density as will be seen below.

The capability for providing electrostatic mass selection within the ionization cell depends, not only on generating all ions at a uniform potential, but also on maintaining the extraction grid at a positive potential relative to the plasma-space potential. When a plasma fills the ionization-cell volume, the plasma space potential reaches an equilibrium value that is fixed relative to the most positive boundary, which would be the extraction grid (i.e., the extraction grid becomes the ionization cell anode in effect). Therefore, for operation at increased plasma density, it was necessary to add an anode potential grid to shield the extraction grid from the ionization-cell plasma so that electrostatic mass separation could be achieved.

Also, for a fixed grid mesh, increasing the plasma density ultimately resulted in the plasma penetration of the gridded boundaries of the ionization cell and, subsequently, the control of ion beam formation was lost. Because of this, it was thought that application of a magnetic field along the direction of electron flow would limit the electron/neutral interaction volume, reduce the ionization of atoms, and confine the plasma to
the ionization cell to a greater degree. While application of this transverse magnetic field did limit the plasma production volume, it also inhibited the plasma drift toward the extraction boundary. Thus, the net effect of the uniform magnetic field was to reduce the ion cluster current.

Based on these measurements, the ion cluster beam configuration was revised. The ionization cell was located farther downstream from the nozzle, and another skimmer was added to reduce the density of gas atoms in the ionization cell. A second ionization cell was designed and built as shown in Figure 7. A photograph of the ionization cell is shown as Figure 8. Some important features of this cluster beam apparatus are: (1) neutral gas density in the ionization cell is at least a factor of four lower than in previous cluster beam experiments, (2) the ribbon emitter is oriented with its long dimension transverse to the beam axis to reduce the electron/neutral interaction volume, (3) the extraction grid, (G3), and the accelerating grid, (G4), have apertures that are matched and aligned to minimize ion impact on the accelerating grid, and (4) the diameter of the extracted ion beam is about twice as large as previously.

The first experiments, performed to evaluate the cluster beam apparatus shown in Figure 7, were a repeat of the measurements of cluster size as a function of gas reservoir pressure with argon gas. The data obtained did not agree with the results reported by Hagen and Obert, as had been observed with the previous cluster beam apparatus. For the new apparatus, the average cluster size was found to be larger than predicted at any given pressure, and also, the cluster size increased more rapidly with pressure than predicted. While exploring all of the possibilities for the differences, it was discovered that a new nozzle had been installed with the new ionization cell, and that the expansion ratio for this new nozzle (about 1300) was higher than that of any nozzle previously documented. Therefore, the
Figure 7. Experimental configuration for second iteration on ion cluster source.
Figure 8. Photograph of the ionization cell that is shown schematically in Figure 7.
neutral gas-flow field measurements described earlier were repeated for this nozzle. The flow from the high-expansion ratio nozzle was found to be much more highly collimated than expected and the gas velocity was only about 80% of the theoretical limiting velocity for supersonic expansion. Moreover, experimental data were less reproducible than for other nozzles.

A nozzle with a throat diameter of 0.254 mm and expansion ratio of 156 was then installed in the ion cluster-beam apparatus configuration of Figure 7. Experiments with this nozzle produced results that were in agreement with previous results and those reported by Hagen and Obert. Figure 9 compares the cluster-size versus reservoir-pressure relationships measured using the nozzles discussed here with the results reported by Hagen and Obert. It should be noted that the scaling relations governing nozzle diameter and reservoir pressure as defined by Hagen and Obert have been used to adjust for the different nozzle-throat diameters.

The next experiments that were performed with the improved ion cluster-beam system explored the effect of ionizing current on the cluster size distribution. The ionization-cell anode was set to 100 V, and the ionization current was adjusted by changing the emitter temperature. The emission current was readily variable from 7 to 20 mA under "standard" operating conditions. Figure 10 compares normalized retarding potential characteristics measured for several values of ionizing current. The collector current refers to the Faraday cup measurement described earlier and the ion cluster beam system was operated with the ion beam voltage set to 500 V. The collector current was therefore normalized to the value measured at 500 V. It is apparent in Figure 10(a) that the cluster size distribution was not affected appreciably by varying the ionizing current within the limits shown (at least for the larger clusters).

In the course of these cluster size distribution determinations, as deduced (Figure 10(b)) from the retarding
Figure 9. Comparison of results for experimental determination of average cluster size versus reservoir pressure. Nozzle scaling relationship of Hagen and Obert$^2$ used to adjust pressure.
a) Retarding potential characteristic for several values of ionizer-emission current.

b) Cluster size distribution deduced from retarding potential characteristics shown in (a).

Figure 10. Retarding potential measurement characteristics for several values of ionizer emission current.
potential characteristics of Figure 10(a), some anomalies were noted. Ideally, the collector current, measured as a function of increasing retarding potential, should be constant up to the potential at which the ions are generated, if all ions are generated at a uniform potential. The explanation for the variation in current seen in this region of the current characteristic is under investigation at this time. Some possibilities that have been explored up to this point include:

- Current contributions from charge exchange ions
- Current contributions from cluster ion fragments that have broken apart because of gas collisions in the ion drift region (downstream of the ion extraction grids)
- Ion focusing effects in the Faraday cup
- Secondary electron currents in the Faraday cup
- Nonaxial velocity components in cluster ion trajectories

The collector current variations, as seen in Figure 10(a) in the 100 V retarding potential region are thought to be caused by focusing and secondary electron effects in the gridded Faraday cup. The overall increase in collector current over the value at 500 V is at least partially due to nonaxial components in the cluster ion trajectories. There are experimental inferences that some of the current drop measured at retarding potential values below 500 V may be the result of fragmented clusters or atomic ions generated in the acceleration region.

Even though the Faraday probe measurements suffer from some inaccuracy, it is possible to estimate a few of the operating characteristics of the ionization cell for typical conditions. Based on the measured gas-flow characteristics, the density of neutral gas atoms in the ionization cell is approximately $3 \times 10^{13}$ per cm$^3$. Estimating one-third of these to be clustered, with an average size of 2000 atoms/cluster, the cluster density in the ionization cell is $5 \times 10^9$ clusters/cm$^3$. Current measured in the
cluster ion beam extrapolates to $6.4 \times 10^8$ cluster ions/cm$^3$ at the extraction grid, or about 13% ionization (see Table I). Although these quantities are only inferred from measured quantities, they are considered sufficiently accurate to characterize the ion cluster-beam system for the purpose of evaluating cluster assisted deposition and etching processes.

Table I. Typical Operating Conditions for the Improved Ionization Cell

<table>
<thead>
<tr>
<th>Ionizer Voltage</th>
<th>100 V</th>
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<tr>
<td>Ionizer Current</td>
<td>20 mA</td>
</tr>
<tr>
<td>Extraction Voltage</td>
<td>2000 V</td>
</tr>
<tr>
<td>Extracted Ion Current</td>
<td>8 µA</td>
</tr>
<tr>
<td>Average Cluster Size</td>
<td>2000 atoms</td>
</tr>
<tr>
<td>Cluster Density (Neutral)</td>
<td>$5 \times 10^9$ cm$^{-3}$</td>
</tr>
<tr>
<td>Cluster Density (Ions)</td>
<td>$6.4 \times 10^8$ cm$^{-3}$</td>
</tr>
<tr>
<td>Ionization Fraction</td>
<td>13%</td>
</tr>
</tbody>
</table>

Ionization Cells for Nonvolatile Material Clusters

In the initial cluster beam experiments at Hughes, the current density distribution across the ionized beam was found to be very nonuniform and it was concluded that nonuniformity in the space potential within the ionization cell was the most probable cause. In fact, it is to be expected that injection of the ionizing electron current into the ionization cell results in a large negative space charge, and this space charge will produce a parabolically shaped depression in the space potential within the ionization cell as illustrated in Figure 11. The depression on the axis can be determined from
Figure 11. Potential distribution in ionization cells operated at low pressures (<10^{-3} Torr) where the electronic space charge is only partially compensated by ionic space charge.
where \( r \) is the cell radius, \( j \) is the average electron current density, and \( v \) is the electron velocity. For relatively small depressions in space potential, the dependence of \( v \) on the depth of depression can be neglected and

\[
v = \sqrt{\frac{2eV_o}{m}}
\]

where \( V_o \) is the anode voltage. Based on these relationships, a depression in space potential of about 50 volts is found for a cell of 0.5 cm radius, operated with a moderately low-electron current density of 30 mA/cm\(^2\). Clearly, ions generated in such a cell originate from different potential levels, and therefore, differ significantly in energy. Also, ions generated off-axis are accelerated towards the axis, and thus, form a beam with large transverse velocity components.

The condition for creating a depression in space potential within the ionization cell as described here, occurs only when the ion production rate is insufficient to balance the electron density generated by the ionization cell emitter. This occurs if the neutral density is very low in the ionization cell (i.e., operation at low pressure), or if the emission current is very high, or both. Both conditions are expected to occur with nonvolatile with cluster beams where the vapor pressure in the ionization cell does not exceed \( 10^{-3} \) Torr. In ionization cells for volatile material clusters, as described in the preceding section, the pressure of clustered and atomic gas is expected to be at least three orders of magnitude greater and formation of a neutral plasma with level potential distribution is assured.

A primary objective under this program was to develop an ionization cell that produces ionization at uniform potential (no depression in space potential). As in the example described
earlier for gas clusters, a small difference in potential levels is acceptable. When working with silver clusters of about 1000 atoms, the tolerable variation is about 5 V since the kinetic energy of such clusters (while neutral) is on the order of about 20 eV.

Early in the investigation, several simplistic approaches were considered and evaluated for obtaining unipotential ionization as illustrated in Figure 12. Compared with the Takagi configuration, Figure 12(a), the approaches shown in Figures 12(b) and (c) use narrowly spaced anode plates inside the ionizer, and place the ionizer in close proximity to the crucible nozzle. Both methods, 12(b) and 12(c), have resulted in some improvement over 12(a); however, they also had some deficiencies. The multiple anode configuration rendered extracted beams even more nonuniform, laterally, and the close-in ionizer was quickly rendered inoperable by deposition of metal vapor on electrodes and insulators.

Fortunately, recent efforts have proven more successful. During a series of tests of a retarding field mass separator, using the configuration shown as Figure 12(d), it was noted that the ions produced had a comparatively narrow energy distribution. This result indicated that the space potential distribution in the ionization cell must have been relatively uniform. The most unusual feature in this operating mode was that the potential level, at which the ions were generated, varied with the potential applied to the retarding grid. This behavior suggested the following ion generation model: As the electron current is turned on, a parabolic space-charge depression forms across the ionization cell. Then, when ions are formed, the bottom of the depression rises up until a level is reached from which ions can escape through the grid openings at a rate which equals the generation rate. In other words, the retarding grid acts as an escape barrier.

A series of experiments has confirmed this model and has resulted in several refinements in understanding. The
Figure 12. Ionizer configurations tested at Hughes.
experimental data is summarized in Figure 13. Figure 13(a) pertains to a low electron current, where the space charge depression is expected to be minimal compared with the cases of Figures 13(b) and (c) which apply to larger emission currents. These figures give the ion current cut-off potentials, as measured with a Faraday cup analyzer, as a function of the potential applied to the retarding grid of the mass separator. The gas pressure serves as a parameter. The vertical bars indicate the range within which the ion current collected by the Faraday cup drops from 90% to 10%.

The data of Figures 13(a), (b), and (c) can be interpreted as follows: at low pressures, where few ions are generated, the space potential level within the cell is very nearly equal to the potential of the mass analyzer grid, implying that the ions can depart readily through this grid once they have raised the space potential to the grid level. At higher pressures, the ion production rate is higher, and the space potential must rise above the grid potential in order that ions can leave in sufficient numbers. At pressures above about $10^{-3}$ Torr, the space potential has risen practically to the level of the anode (200 V in all cases), irrespective of the mass separator potential. The difference between different levels of electron emission is that, with low emission, the potential depression is small and the space potential tends to be high. At the lowest emission levels, Figure 13(a), the space charge depression is negligibly small. In all cases, the basic energy width of the extracted ions falls into the range of 5 to 10 eV, which is quite acceptable. In cases where the space potential stays lower than the anode potential, however, the energy distribution of the extracted ions has a high-energy tail. Figure 14 shows the ion current collected by the Faraday cup as a function of the Faraday cup acceptance potential. The ion current is collected at two energy bands; the majority of current is comprised of ions with a 5 to 10 eV energy spread, and a smaller fraction of current is
Figure 13. Ionizer space potential plateaus and ion generation potential ranges (10 to 90%) in low pressure ionizers with extraction grids.
Figure 14. Ion energy distribution of atomic ions obtained from a low pressure ionizing cell.
collected with an energy spread that corresponds to the voltage difference between the mass separator grid and the anode. These two energy distributions can be explained by an ionization cell space potential distribution that is essentially parabolic, but has a flat bottom in the center of the cell (see Figure 11). The major portion of the ion population is generated in the bottom region of the potential distribution at the center of the cell and the smaller portion is generated in the steeply varying potentials near the boundaries of the cell. It is expected that the magnitude of the sidewall contribution is smaller for higher emission current since the space charge is larger and the sidewalls are steeper. Earlier observations on ionization cells operated without mass separator grid, with just two aperture plates for ion extraction and acceleration, have shown wide ion-energy widths under all but the lowest electron current levels, indicating that ions do not accumulate in the bottom of the parabolic space charge depression, but are extracted quickly. The important finding of this study, therefore, is that ionization cells must be furnished with an extraction (or mass separating) grid and that this grid must be maintained at or above anode potential if ions with a truly narrow energy distribution and good collimation are to be obtained. In fact, as discussed earlier, if the mass separator grid is kept several volts above the anode potential, it retards all ions with less than a few electron volts; i.e., all atomic and small cluster ions.
WORK PLANNED

Work under the Air Force Program will proceed towards the demonstration and evaluation of thin-film deposition using cluster ion beams generated with the newly developed apparatus. Relationships will be sought between ionization cell and mass separator performances and the quality achieved in the deposition processes. Inert-gas cluster-assisted deposition of gold on GaAs substrates will be explored in the existing gas cluster facility. Plans for direct deposition of nonvolatile materials with cluster beams are less certain so far, because a relatively low fraction of clusters has been observed in cluster-source tests at Hughes.
REFERENCES


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