High temperature metal atom beam sources

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This paper presents a survey of the factors governing the performance and operation of high temperature subsupersonic metal atom beam sources. After an initial statement of the requirements placed on such sources a section is presented which considers the factors determining atomic beam intensities and profiles. The section which considers the materials used in source construction discusses the choice of crucible material, and in so doing presents a table of the most suitable materials, hazard assessments, and other information for all those elements which can be vaporized. Two further parts of this section are devoted to resistive heater materials and ceramics. The review of the sources is divided between resistively heated sources, sources heated by electron bombardment, and inductively heated sources. Finally there is a section which briefly discusses the monitoring of source performance. © 1995 American Institute of Physics.

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I. INTRODUCTION

The preparation of free neutral atoms in atomic beams provides an excellent means for many detailed studies of their properties and their interaction with particles and fields. Atomic beam experiments comprise atom–atom, atom–ion, atom–electron, atom–photon scattering, the magnetic resonance methods, photoelectron and fluorescence spectroscopy, and atom interferometry. In addition the interaction of an atomic beam with electromagnetic fields provides extremely precise frequency standards. Recently the scattering of polarized electrons and their interaction with laser and vacuum ultraviolet light has provided very deep insight into many-electron dynamics.

In principle, the construction of an atomic beam source is simple. It requires a container in which a certain vapor pressure of the substance under study is sustained, a well-defined exit opening, and a set of apertures. However, in actual practice, especially for metal atoms, many problems arise in the construction of a suitable container, the heating element, and the orifice. The problems vary from element to element and therefore one often has to resort to quite different approaches, materials, and designs.

In this article, we have tried to collect the information that is available on metal atom beam sources. The emphasis is on providing the basis for the reader to develop the optimal atomic beam source for his purpose. In Sec. II we will discuss the different requirements and constraints. The basic laws governing the intensity and angular distribution of the atomic beam are presented in Sec. III. The choice of the right material is crucial for the successful operation of a metal atom beam source. Section IV focuses on these materials problems. In Sec. V the main types of metal atom beam sources are presented and discussed. Monitoring of source performance is the content of Sec. VI.

II. REQUIREMENTS AND CONSTRAINTS

A. Introduction

The requirements and constraints which a high temperature metal atom beam source must satisfy depend on the particular experiment or application being considered. In this article we will focus our attention on the investigation of the
interaction of free metal atoms with photons or charged particles, e.g., electrons or protons, the quantities that can be determined in such an experiment being the number, energy, angular distribution, and polarization of the outgoing photons, electrons, atoms, and ions. In this section we will give a brief discussion of the most important characteristics required in atomic beams which are to be used for such investigations.

B. Cross section of the atomic beam

The cross section of the atomic beams is determined by the cross section of the photon or particle beam and by the requirements set by the detectors of the outgoing particles in respect to energy and angular resolution. Typical beam diameters for circular cross sections vary between 0.1 mm and approximately 1 mm.

C. Density of atoms in the interaction region

The typical atom densities range from $10^8$ up to $10^{13}$ atoms/cm$^3$, this upper limit being determined by the capability of the source itself and the usual requirement that the effects of collisions between the atoms are negligible. If the alignment or orientation of the atoms generated by, for example, laser excitation has to be conserved, the maximum densities affordable lie generally in the order of $\leq10^{10}$ atoms/cm$^3$, this limit being set principally by radiation trapping and low energy electron scattering. The lower limit for the atomic density is determined by the signal to background ratio which in turn depends on the interaction cross sections and the properties (angular acceptance, transmission, detection efficiency) of the detectors.

D. Collimation of the atomic beam

In order to achieve high atomic densities it is advantageous to position the orifice of the oven very close to the interaction region. However, difficulties can arise from this arrangement due to poor collimation of the beam, electrons and ions emanating from the hot orifice, thermal radiation, and electric and magnetic stray fields generated by the oven. Increasing the distance between the oven and the interaction region reduces these detrimental effects and allows for good collimation of the beam by apertures, collimation being essential to reduce Doppler broadening in, for example, laser pumping between hyperfine levels. On the other hand, increasing the distance reduces the density of atoms in the interaction region; for example, Anderson et al. found that for a beam with an angular spread of 1 mrad achieved by 1-mm-diam apertures separated by 1 m the density is less than $10^9$ atoms/cm$^3$. A well-collimated beam of metal atoms reduces the problems arising from metal contamination of the vacuum system, the analyzers, and detectors. The beam can be trapped on a cooled beam stop, which also greatly facilitates a safe disposal of the material. In addition there is always the problem of clogging of apertures which in many cases can only be overcome by heating and all its negative consequences, such as high radiation losses by unshielded apertures, thermal emission of electrons, and associated stray fields.

E. Stability of the atomic beam

The number of electrons, ions, and photons registered by the detectors in any crossed-beam experiment depends directly on the target density. Stability of the atomic beam in time and space is therefore crucial. When the atomic source has been raised to operating temperature and has reached equilibrium it should be able to operate under stable conditions for at least several hours. It is advisable, initially, to raise the temperature of the empty furnace slowly and carefully to a temperature slightly in excess of the operating temperature required for the material to be used if spurious signals are to be avoided. Samples, especially new ones, have to be outgassed, if possible before reaching their operating temperature, if beam instabilities are to be minimized; dramatic instabilities may occur upon melting and boiling which can result in the ejection of lumps of material from the oven aperture. For atoms prepared at very high temperatures materials problems, for example, alloying, often seriously interfere with the efforts to achieve a stable beam. Further, the metal reservoir of the beam source must contain sufficient metal for prolonged operation; limits set by space, safety, and materials requirements make periodic recharging of the furnace unavoidable. Clogging of the apertures and contamination of the surroundings are frequently further reasons for interrupting the measurements and opening the system. Therefore recharging and cleaning has to be taken into account in the design of the beam source. It should be easily removable and rechargeable, and in an ideal system the alignment of the atomic beam should not be affected by these operations. If this is not possible alignment should be achievable in a reasonable time.

F. Compatibility with the system

Usually the atomic beam source is mounted in a high vacuum or ultrahigh vacuum system. The source therefore has to meet the vacuum conditions. The materials used should have low outgassing rates at elevated temperatures and the structure should provide apertures sufficient for effective pumping. Heat transfer to the environment has to be kept to a minimum. Contamination of the vacuum system and the particle analyzers and detectors by metal vapor deposition has also to be minimized. Stray thermal electrons, atoms, and ions originating from the beam source raise the background count rate unless they are prevented from reaching the detectors.

Electric and magnetic fields created by the heating currents can seriously distort the path of charged particles. By careful wiring and electric and magnetic shielding such fields can be minimized.

III. INTENSITY AND ANGULAR DISTRIBUTION

It is the objective of this article, not only to describe types of furnaces which may be used to produce atomic beams, but also to pay some attention to the quality of the beams so produced. This latter parameter involves a discussion of both the rate at which material leaves the furnace, and also the profile or directionality of the resulting atomic beam.
Directionality in atomic beams is achieved by a combination of the selection of a suitable oven aperture and operation in a regime where the Knudsen number ($K_n$), the ratio of the mean free path of the source atoms to the diameter of the aperture of the oven, is greater than unity. Under these conditions there is molecular effusion from the source. This condition generally holds for metal vapor pressures of less than $10^{-1}$ mm Hg and oven apertures of up to 2 mm diam. The mean free path is given by $\lambda = \frac{7.321 \times 10^{20}T}{P \sigma}$ cm, where $T$ (K) is the temperature, $P$ (mm Hg) is the source pressure, $\sigma$ in cm$^2$ is the molecular collision cross section. Under molecular effusion conditions the angular distribution of atoms or molecules effusing through a thin aperture source at an angle $\theta$ into a solid angle $d\omega$ is a cosine distribution $dN = \frac{d\omega}{4\pi} n \bar{v} B \cos \theta$ atoms/s, atoms/cm$^2$, where $R$ is the source area (cm$^2$), $n$ is the particle density in the source (atoms/cm$^3$), and $\bar{v}$ is the mean molecular velocity (cm/s) of the atoms or molecules in the source and is given by $\bar{v} = 1.4551 \times 10^4 \left( \frac{T}{M} \right)^{1/2}$ cm/s, where $M$ is the atomic weight in grams.

Extending these general statements from the "ideal" thin aperture to the more practical case of a tube of length $L$ cm, diameter $a$ cm, where $L/a = 1$, the flow of atoms through such a tube is said to be transparent, or there is said to be molecular effusion, when the mean free path is greater than the length of the tube. Under this condition the beam is highly directional, the angular spread being given by the dimensions of the aperture, and the peak intensity $I$ (atoms/s/sr) and flow rates $N$ (atoms/s) are proportional to the pressure in the source.

The condition of hydrodynamic flow, where the mean free path is less than the aperture diameter is not of interest to the present article.

Of more practical use, because it allows higher source pressures and hence greater beam intensities, is the condition where the mean free path is greater than the diameter of the tube and less than the length. In this regime an increase of pressure results in an increased beam width and intensity; as an example, peak intensities of the order $10^{-12}$ atoms/s can only be achieved with source pressures greater than $10^{-1}$ mm Hg where the mean free path is of the order $10^{-2}$ cm.

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Extending these general statements from the "ideal" thin aperture to the more practical case of a tube of length $L$ cm, diameter $a$ cm, where $L/a = 1$, the flow of atoms through such a tube is said to be transparent, or there is said to be molecular effusion, when the mean free path is greater than the length of the tube. Under this condition the beam is highly directional, the angular spread being given by the dimensions of the aperture, and the peak intensity $I$ (atoms/s/sr) and flow rates $N$ (atoms/s) are proportional to the pressure in the source.

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multichannel array. Commercially available multichannel arrays are also generally fabricated from stainless steel.

Calculation of the mean free path depends on reliable data for the molecular cross section. Such data have usually been deduced from viscosity or diffusion measurements, in addition to a number of experiments where the cross section is directly obtained from measurement of the attenuation of a beam of atoms on passing through a collision cell.

Although this latter method leads directly to a value of the mean free path, and hence the molecular cross section, very little work has been published on the scattering of a metal vapor atom by its own vapor; there are the data of Estermann et al. on Cs–Cs cross sections and the work of Buck and Pauly on Na, K, Rb, and Cs where cross sections have been measured for collisions between like atoms and all other pair combinations of the group. Both sets of data for Cs–Cs collisions are in excellent agreement, perhaps serendipitously.

In order to calculate the intensity for any given oven arrangement it is necessary to know, not only the vapor pressure corresponding to the temperature of the charge, but also the pressure at the oven aperture. Motzfeldt discussed this problem for the oven arrangement shown in Fig. 2, which shows a cylindrical oven with a thin aperture. By considering the pressure at two planes, 1 and 2, an expression was obtained for the pressure at plane 2, that is in the region of the aperture, in terms of the pressure at plane 1 and a number of geometrical factors

$$P_2 = P_1 \left(1 + \frac{1}{\alpha + \frac{1}{WA} - 2}\right) \frac{PW_B}{A},$$  

where $P$ is the pressure at plane 2, $P_1$ is the pressure at plane 1, $\alpha$ is the accommodation coefficient, which can be assumed to be approximately unity,$^{12}$ $W_A$ is the Clausing probability factor for the oven itself,$^{13,14}$ $W_B$ is the Clausing factor for the aperture, and $A$ and $B$ are the areas of the cross section of the oven and aperture, respectively; the Clausing probability factor is the probability that an atom enters the aperture and goes through it without having been back into the original chamber; values of Clausing factors have been tabulated by Dushman.$^{14}$

For an oven where the flow through the aperture is transparent the axial intensity at a distance $d$ is given by Ramsey$^2$ [Eq. (II.16) of Ref. 2] as

$$I = \frac{1.118 \times 10^{22} PB}{d^2(MT)^{1/2}} \text{ atoms/s/cm}^2. \quad (5)$$

This expression is in fact identical to the peak-intensity equation, Eq. (2), for an ideal aperture of zero length.

For the case of transparent flow through a tube aperture, length $L$, diameter $a$, the beam width at half-maximum intensity is$^5$ [Eq. (19) of Ref. 5]

$$\theta_{1/2} = 1.68(a/L). \quad (6)$$

Very often in scattering experiments where the oven aperture is in the form of a tube, cross sections for the processes being studied are too small to allow the luxury of working with atomic beams which are transparent. As the pressure of such a source is increased such that the mean free path is less than the length of the aperture but still greater than the diameter, the peak intensity of the beam becomes proportional to the square root of the pressure and the beam width at half-maximum is proportional to the square root of the total flow rate. The peak intensity is then given by$^5$ [Eq. (12) of Ref. 5]

$$I = 7.3 \times 10^{12} \frac{B}{\sigma} \left(\frac{P}{\sigma LM}\right)^{1/2} \text{ atoms/s/cm}^2 \quad (7)$$

and the beam width at half-maximum intensity is given by$^5$ [Eq. (27) of Ref. 5]

$$\theta_{1/2} = 8.3 \times 10^5 a \left(\frac{\sigma P}{LT}\right)^{1/2}. \quad (8)$$

Once $K_a$ becomes less than unity (but the beam is not supersonic) further increases in the source temperature do not increase the peak intensity; interatomic collisions near the exit of the aperture result in a gas cloud around this region which effectively increases the pressure outside the aperture and reduces effusion from the source.

It is worth noting that in this high pressure regime the characteristics of the beam are determined by the final section of the aperture tube. The length of the tube only serves to reduce the pressure, so that where operation in the viscous mode cannot be avoided a long tube aperture should not be used; in such circumstances a long tube will necessitate use of unwarranted high furnace temperatures and correspondingly high pressures.

Hanes$^{15}$ has demonstrated that for a tube aperture being operated such that the Knudsen number $K_a$ is close to unity at the exit, and therefore having conditions of viscous flow at the entrance, the effective length of the aperture where molecular flow occurs is the end section. Taking as an example the condition where $K_a = 1$ the “effective length” of the aperture is then given by $L_e = \lambda$. When this expression for $L_e$ is substituted into Eq. (7) the expression for the axial intensity differs from Eq. (7) by being only a factor 0.8 smaller, a difference which arises from the “precollimation” prior to reaching the region of molecular flow.
TABLE I. Resistivities (Ref. 17) and skin depths (at 1 MHz) for a number of materials at 20 °C.

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>Ti</th>
<th>Mo</th>
<th>Ta</th>
<th>W</th>
<th>SS*</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$ ((\mu\Omega) m)</td>
<td>13.7</td>
<td>0.42</td>
<td>0.05</td>
<td>0.12</td>
<td>0.05</td>
<td>0.73</td>
</tr>
<tr>
<td>$\delta$ (mm)</td>
<td>1.85</td>
<td>0.33</td>
<td>0.12</td>
<td>0.18</td>
<td>0.12</td>
<td>0.43</td>
</tr>
</tbody>
</table>

*304 stainless steel.

IV. MATERIALS FOR USE WITH HIGH TEMPERATURE ATOM BEAM SOURCES

A. Crucibles

The choice of crucible for containing a metal which is to be evaporated is determined by the heating technique which has been selected and, even more important, the compatibility of the hot metal and the crucible material. Solubility and reactivity problems become extreme at high temperatures where molten liquid metals are being contained; crucibles which, for example, are suitable for molecular beam epitaxy (MBE) evaporation sources, working at temperatures equivalent to vapor pressures of \(10^{-4}\) mm Hg can become quite unsuitable at the temperatures which are required to achieve vapor pressures of \(10^{-2}\) mm Hg. Metals which require only low temperatures for their evaporation can become surprisingly aggressive to materials in which all other respects appear unreactive. For example, we have found a stainless steel oven to fail when evaporating zinc at only 350 °C; the oven separated into two parts at the level of the liquid zinc due to liquid metal embrittlement and the formation of a low temperature eutectic which apparently melted at this temperature. Similarly, at 1000 °C lithium forms a low melting point alloy with stainless steel.\(^{12}\) Phase diagrams\(^{16}\) might be expected to be an important resource in choosing crucible materials. However, many published phase diagrams are inaccurate and such diagrams should be viewed with great caution.

In the case of metals which are to be evaporated by induction heating a second constraint applies to the choice of crucible if the crucible is also to be the susceptor. This is the resistivity. Table I lists the resistivities at 20 °C of a number of useful materials for use as both susceptor and crucible.\(^{17}\) By far the best of these from a heating point of view is carbon with its very high resistivity, although as can be seen from Fig. 3, which plots resistivity as a function of temperature for a number of elements,\(^{18}\) carbon has a minimum resistivity of 6.4 \(\mu\Omega\) m at 1000 °C while the resistivity of the metals generally increases with temperature; the difficulty of induction heating tungsten, for example, decreases with increasing temperature.

Carbon in the form of high density graphite is a suitable crucible material for a wide range of metals. High density graphite, which machines very easily and has good mechanical strength, is available in a number of different grades to suit different applications. Manufacturers such as Ringsdorf are able to provide advice for specific applications; a wide range of high density graphite is available.

Crucibles can be manufactured from refractory metals in the laboratory without difficulty using spark erosion techniques. However, it is worth noting that Metallwerk Plansee can supply machined parts in refractory metals, and Ultramet are able to produce refractory metal crucibles using chemical vapor deposition. A limited range of refractory metal crucibles is available from Cerac.

All the common insulators such as alumina, zirconia, boron nitride, and berillia, are excellent crucible materials, but having resistivities greater than \(10^7\) \(\mu\Omega\) cm they cannot be used as susceptors for induction heating. A comprehensive range of hot pressed, binder-free, crucibles manufactured from a range of insulating materials is available from Cerac. Further discussion of machinable ceramic materials is given in Sec. IV C below.

Table II is a list of as many sources as we could identify which have been used to produce beams of elements. We do not expect such a list to be complete, but it is a serious attempt to present the state of the subject. In many cases more than one type of crucible is suggested for a particular element. Before deciding upon a particular crucible material it is important to refer to the original papers cited in order to ascertain whether the crucible material suits the particular experiment being considered. With the exception of U, we have not mentioned radioactive sources.

We have taken some care to comment in Table II on the hazards which we are aware of in connection with the handling of elements. This danger is often overlooked, and experimenters are well advised to treat the question of potential hazards associated with handling certain elements very seriously. While we have attempted to list the hazards associated
<table>
<thead>
<tr>
<th>Element</th>
<th>Melting point (^{b}) (°C)</th>
<th>Boiling point (^{b}) (°C)</th>
<th>Temperature for vapor pressure of (10^{-2}) mm Hg (°C)</th>
<th>Crucible material</th>
<th>Ref.</th>
<th>Hazards</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminum</td>
<td>659</td>
<td>2467</td>
<td>1220</td>
<td>TiB(_2), BN, BN, W</td>
<td>19–22</td>
<td>2</td>
<td>Wets all materials, creeps, alloys with W, forms nitride with BN, reacts with ceramics.</td>
</tr>
<tr>
<td>Antimony</td>
<td>630</td>
<td>1440</td>
<td>540</td>
<td>C, quartz</td>
<td>23,24</td>
<td>3</td>
<td>Dimer vapor</td>
</tr>
<tr>
<td>Sb(_4)</td>
<td>310</td>
<td>540</td>
<td>1220</td>
<td>C, quartz</td>
<td>23,24</td>
<td>3</td>
<td>Dimer vapor</td>
</tr>
<tr>
<td>Arsenic</td>
<td>820</td>
<td>610</td>
<td>310</td>
<td>C</td>
<td>25,26</td>
<td>4,5</td>
<td>Dimer vapor</td>
</tr>
<tr>
<td>As(_2)</td>
<td>310</td>
<td>610</td>
<td>310</td>
<td>C</td>
<td>25,26</td>
<td>4,5</td>
<td>Dimer vapor</td>
</tr>
<tr>
<td>Barium</td>
<td>710</td>
<td>1770</td>
<td>690</td>
<td>Metals, SS, Ta</td>
<td>27,28</td>
<td>1</td>
<td>Wets most metals without alloying. Reacts with ceramics.</td>
</tr>
<tr>
<td>Bismuth</td>
<td>271</td>
<td>1530</td>
<td>675</td>
<td>Metals, C</td>
<td>25,32</td>
<td>2,7</td>
<td>Wets full crucible</td>
</tr>
<tr>
<td>Bi(_2)</td>
<td>271</td>
<td>1530</td>
<td>675</td>
<td>Metals, C</td>
<td>25,32</td>
<td>2,7</td>
<td>Wets full crucible</td>
</tr>
<tr>
<td>Boron</td>
<td>2030</td>
<td>2550</td>
<td>2100</td>
<td>C</td>
<td>24,33,34</td>
<td>1,3</td>
<td></td>
</tr>
<tr>
<td>Cadmium</td>
<td>321</td>
<td>767</td>
<td>260</td>
<td>Metals, SS</td>
<td>35–38</td>
<td>2,3,5</td>
<td>Low sticking coefficient spoils vacuum system.</td>
</tr>
<tr>
<td>Caesium</td>
<td>28</td>
<td>669</td>
<td>150</td>
<td>SS, Ta</td>
<td>39–42</td>
<td></td>
<td>Reacts violently in air, creeps, low sticking coeff., deteriorates detectors.</td>
</tr>
<tr>
<td>Calcium</td>
<td>850</td>
<td>1440</td>
<td>600</td>
<td>Metals, SS, Al(_2)O(_3), V4A</td>
<td>43–47,80</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Carbon</td>
<td>3500</td>
<td>3900</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cerium</td>
<td>700</td>
<td>3426</td>
<td>1750</td>
<td>Mo, Ta</td>
<td>27,48</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Chromium</td>
<td>1850</td>
<td>2600</td>
<td>1400</td>
<td>Zirconia lined (C, Mo), Al(_2)O(_3), Ta, Sub</td>
<td>44,49–57</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>Cobalt</td>
<td>1405</td>
<td>7000</td>
<td>1530</td>
<td>Zirconia lined (Ta,Mo), Al(_2)O(_3)</td>
<td>44,57–56</td>
<td>3,5</td>
<td>Alloys with refractory metals</td>
</tr>
<tr>
<td>Copper</td>
<td>1083</td>
<td>2580</td>
<td>1250</td>
<td>C, Al(_2)O(_3), W Ta, Mo</td>
<td>49,51,64,64</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Dysprosium</td>
<td>1412</td>
<td>2300</td>
<td>1130</td>
<td>W</td>
<td>77</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Erbium</td>
<td>1530</td>
<td>2600</td>
<td>1250</td>
<td>Ta</td>
<td>65</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Europium</td>
<td>326</td>
<td>1440</td>
<td>620</td>
<td>C, W, Mo</td>
<td>27,53,66–68</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gadolinium</td>
<td>1312</td>
<td>2700</td>
<td>1600</td>
<td>Alumina lined Ta, W</td>
<td>27</td>
<td>2</td>
<td>Alloys with Ta</td>
</tr>
<tr>
<td>Gallium</td>
<td>958</td>
<td>2880</td>
<td>1400</td>
<td>C, Ta, Al(_2)O(_3)</td>
<td>69–71</td>
<td></td>
<td>Alloys with refractory metals</td>
</tr>
<tr>
<td>Germanium</td>
<td>958</td>
<td>2880</td>
<td>1400</td>
<td>C, W</td>
<td>25,72,73</td>
<td>2</td>
<td>West Ta, Mo</td>
</tr>
<tr>
<td>Element</td>
<td>Melting point (°C)</td>
<td>Boiling point (°C)</td>
<td>Temperature for vapor pressure of 10⁻² mm Hg (°C)</td>
<td>Source</td>
<td>Cruible material</td>
<td>Ref.</td>
<td>Hazards</td>
</tr>
<tr>
<td>---------</td>
<td>------------------</td>
<td>------------------</td>
<td>----------------------------------</td>
<td>--------</td>
<td>----------------</td>
<td>-----</td>
<td>----------</td>
</tr>
<tr>
<td>Gold</td>
<td>1063</td>
<td>2660</td>
<td>1420</td>
<td></td>
<td>C, Mo</td>
<td>25,49,74,75</td>
<td>Wets Mo</td>
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<tr>
<td>Hafnium</td>
<td>2000</td>
<td>3100</td>
<td>2450</td>
<td></td>
<td>W (12% sol. at 2450)</td>
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<tr>
<td>Holmium</td>
<td>1300</td>
<td>2300</td>
<td>1180</td>
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<td></td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Indium</td>
<td>156</td>
<td>2000</td>
<td>940</td>
<td></td>
<td>C, Ta</td>
<td>69,76,77</td>
<td>Wets W, Cu</td>
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<tr>
<td>Iridium</td>
<td>2443</td>
<td>2550</td>
<td></td>
<td></td>
<td>W</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>Iron</td>
<td>1539</td>
<td>2900</td>
<td>1500</td>
<td></td>
<td>Zirconia lined Mo, Al₂O₃</td>
<td>44,52–54</td>
<td>Alloys with all refractory metals</td>
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<tr>
<td>Lanthanum</td>
<td>920</td>
<td>4200</td>
<td>1760</td>
<td></td>
<td>Ta</td>
<td>27,79</td>
<td>Deposit burns in air if scraped</td>
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<td>Lead</td>
<td>327</td>
<td>1750</td>
<td>720</td>
<td></td>
<td>Metal, Ta, Mo</td>
<td>77,81</td>
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<td>Lithium</td>
<td>180</td>
<td>1330</td>
<td>335</td>
<td></td>
<td>33, Mo, TZ–Mo</td>
<td>64,68,82–85</td>
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</tr>
<tr>
<td>Lucentium</td>
<td>1700</td>
<td>1900</td>
<td>1700</td>
<td></td>
<td>Ta</td>
<td>38</td>
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<tr>
<td>Magnesium</td>
<td>650</td>
<td>1100</td>
<td>423</td>
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<td>Metal, C, Sub</td>
<td>86,87</td>
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<tr>
<td>Manganese</td>
<td>1250</td>
<td>2100</td>
<td>960</td>
<td></td>
<td>W, Mo, Ta, C Al₂O₃, Sub</td>
<td>25,44,51,52, 57,61,88,89</td>
<td>Wets refractory metals</td>
</tr>
<tr>
<td>Mercury</td>
<td>–38.9</td>
<td>356.6</td>
<td>48</td>
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<tr>
<td>Molybdenum</td>
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<td>4600</td>
<td>2500</td>
<td></td>
<td>W, Sub</td>
<td>29</td>
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<tr>
<td>Neodymium</td>
<td>1024</td>
<td>3170</td>
<td>1350</td>
<td></td>
<td>W</td>
<td>27</td>
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<tr>
<td>Nickel</td>
<td>1453</td>
<td>2820</td>
<td>1500</td>
<td></td>
<td>Zirconia lined Mo, Al₂O₃</td>
<td>44,52,54,59</td>
<td>Alloys with refractory metals</td>
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<tr>
<td>Niobium</td>
<td>2420</td>
<td>5100</td>
<td>2700</td>
<td></td>
<td>W</td>
<td>92</td>
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<tr>
<td>Nobelium</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Osmium</td>
<td>3045</td>
<td>5027</td>
<td>2950</td>
<td></td>
<td></td>
<td>93</td>
<td></td>
</tr>
<tr>
<td>Palladium</td>
<td>1552</td>
<td>3140</td>
<td>1480</td>
<td></td>
<td>C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Platinum</td>
<td>1772</td>
<td>3827</td>
<td>2100</td>
<td></td>
<td>C, Zirconia lined Mo</td>
<td>25,93</td>
<td></td>
</tr>
<tr>
<td>Plutonium</td>
<td>641</td>
<td>3232</td>
<td>1525</td>
<td></td>
<td>W</td>
<td>94</td>
<td></td>
</tr>
<tr>
<td>Polonium</td>
<td>254</td>
<td>960</td>
<td>320</td>
<td></td>
<td></td>
<td>32</td>
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</tr>
<tr>
<td>Potassium</td>
<td>63.6</td>
<td>774</td>
<td>214</td>
<td></td>
<td>Metals, SS</td>
<td>43,90,95</td>
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<tr>
<td>Praseodymium</td>
<td>931</td>
<td>3312</td>
<td>1400</td>
<td></td>
<td>Thoria lined Mo, W</td>
<td>27,96</td>
<td></td>
</tr>
<tr>
<td>Promethium</td>
<td>1080</td>
<td>2460</td>
<td>1130</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
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<td>Protactinium</td>
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<td></td>
</tr>
<tr>
<td>Radium</td>
<td>700</td>
<td>1140</td>
<td>550</td>
<td></td>
<td>Sub</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rhenium</td>
<td>3180</td>
<td>5630</td>
<td>3050</td>
<td></td>
<td>Sub</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Rhodium</td>
<td>1966</td>
<td>2730</td>
<td>2040</td>
<td></td>
<td>C, Zirconia lined W</td>
<td>25,93</td>
<td></td>
</tr>
<tr>
<td>Rubidium</td>
<td>39</td>
<td>688</td>
<td>175</td>
<td></td>
<td>Cu, metals, SS</td>
<td>96–98</td>
<td>Reacts violently in air</td>
</tr>
<tr>
<td>Ruthenium</td>
<td>2400</td>
<td>3000</td>
<td>2350</td>
<td></td>
<td>Sub</td>
<td>2</td>
<td></td>
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</table>
TABLE II. (Continued.)

<table>
<thead>
<tr>
<th>Element</th>
<th>Melting point&lt;sup&gt;a&lt;/sup&gt; (°C)</th>
<th>Boiling point&lt;sup&gt;b&lt;/sup&gt; (°C)</th>
<th>Temperature for vapor pressure of 10&lt;sup&gt;-2&lt;/sup&gt; mm Hg (°C)</th>
<th>Source</th>
<th>Crucible material</th>
<th>Ref.</th>
<th>Hazards</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Samarium</td>
<td>1077</td>
<td>1791</td>
<td>740</td>
<td></td>
<td>Sub, Ta, Mo</td>
<td>27,53,65,68,88</td>
<td>2</td>
<td>Slight alloying with W</td>
</tr>
<tr>
<td>Scandium</td>
<td>1541</td>
<td>2831</td>
<td>1380</td>
<td></td>
<td>W, (AlO&lt;sub&gt;3&lt;/sub&gt;), (W) , Mo, Sub</td>
<td>44,99–101</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Selenium</td>
<td>217</td>
<td>685</td>
<td>290</td>
<td></td>
<td>Mo, Ta, C, Al</td>
<td>25,93</td>
<td>3</td>
<td>Spoils vacuum system reacts violently in air forms molecules Se&lt;sub&gt;2&lt;/sub&gt;, Se&lt;sub&gt;3&lt;/sub&gt;, Se&lt;sub&gt;7&lt;/sub&gt;</td>
</tr>
<tr>
<td>Silicon</td>
<td>1410</td>
<td>2355</td>
<td>1620</td>
<td></td>
<td>Boron nitride lined C, C</td>
<td>72,73</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Silver</td>
<td>962</td>
<td>2212</td>
<td>1030</td>
<td></td>
<td>C, Mo, Ta</td>
<td>25,57,102–105</td>
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<td></td>
</tr>
<tr>
<td>Sodium</td>
<td>98</td>
<td>883</td>
<td>290</td>
<td></td>
<td>Metals, SS</td>
<td>90,106,107</td>
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<tr>
<td>Strontium</td>
<td>769</td>
<td>1384</td>
<td>530</td>
<td></td>
<td>Metals, SS</td>
<td>97,108,109</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tantalum</td>
<td>2996</td>
<td>550</td>
<td>3050</td>
<td></td>
<td>Mo, Ta, C</td>
<td>25</td>
<td>1.4</td>
<td>Wets metals without alloying</td>
</tr>
<tr>
<td>Tellurium</td>
<td>449</td>
<td>990</td>
<td>370</td>
<td></td>
<td>W</td>
<td>27</td>
<td>2</td>
<td>Wets metals without alloying, forms dimers</td>
</tr>
<tr>
<td>Thulium</td>
<td>1356</td>
<td>3123</td>
<td>1330</td>
<td>W</td>
<td></td>
<td>271</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Thallium</td>
<td>304</td>
<td>1460</td>
<td>630</td>
<td>SS, metals</td>
<td></td>
<td>17,110</td>
<td>5</td>
<td>Wets metals without alloying</td>
</tr>
<tr>
<td>Thorium</td>
<td>1750</td>
<td>4790</td>
<td>2400</td>
<td></td>
<td>Ta</td>
<td>28,68,111</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Thallium</td>
<td>1545</td>
<td>1947</td>
<td>840</td>
<td>Sub, W, Mo</td>
<td></td>
<td>112,113</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Tin</td>
<td>232</td>
<td>2270</td>
<td>1250</td>
<td></td>
<td>C, Ta</td>
<td>25,44,67,</td>
<td>2</td>
<td>Reacts with refractory metals</td>
</tr>
<tr>
<td>Titanium</td>
<td>1660</td>
<td>3287</td>
<td>1740</td>
<td>C, Thoria lined</td>
<td></td>
<td>99,101,114</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Tungsten</td>
<td>3410</td>
<td>5660</td>
<td>3250</td>
<td></td>
<td>Sub</td>
<td>25,99,100</td>
<td>2</td>
<td>Wets Mo without alloying, Slight alloying with W.</td>
</tr>
<tr>
<td>Uranium</td>
<td>1132</td>
<td>3818</td>
<td>2080</td>
<td>W</td>
<td></td>
<td>25,99,100</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Vanadium</td>
<td>1890</td>
<td>3380</td>
<td>1850</td>
<td>UC&lt;sub&gt;2&lt;/sub&gt;/UC mixture in W</td>
<td></td>
<td>25,99,100</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Ytterbium</td>
<td>819</td>
<td>1194</td>
<td>470</td>
<td>Ta, Mo, boron nitride, Sub</td>
<td>68,118–120</td>
<td>2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yttrium</td>
<td>1552</td>
<td>3238</td>
<td>1650</td>
<td>W (based on Se and La)</td>
<td></td>
<td>25,47,93</td>
<td>2</td>
<td>Spoils vacuum system</td>
</tr>
<tr>
<td>Zinc</td>
<td>420</td>
<td>907</td>
<td>340</td>
<td>Ti; metals; Sub, SS*</td>
<td></td>
<td>25,47,93</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Zirconium</td>
<td>1852</td>
<td>4377</td>
<td>2450</td>
<td></td>
<td>W</td>
<td>25,47,93</td>
<td>2</td>
<td></td>
</tr>
</tbody>
</table>

Key to Sources

| Metals         | W, Mo, Ta | 1    | Flammable solid |
| Sub            | Sublimes  | 2    | Flammable powder |
| C              | High density graphite | 3 | Toxic |
| SS             | Stainless steel | 4 | Highly Toxic |
| SS*            | Used by Refer. 47 to 290 °C | 5 | Cancer suspect agent |
| TZ–Mo          | Ti, Zr, C—alloyed Mo | 6 | Harmful in contact with skin |
| V4A            | Nonmagnetic steel | 7 | Harmful vapor |
|                |           | 8    | Radioactive |

Hazard Key (Refs. 122–124)

B. Resistive heater materials

The choice of a heating element for a resistively heated metal vapor beam source is determined primarily by the temperature which is required to be achieved. 1000 °C provides a convenient break point between the use of pure metal wire heaters for achieving higher temperatures and the use of re-
The most important resistance alloys available are the 80/20 Ni/Cr alloys marketed under such names as Nichrome 80, and the range of Kanthal alloys. Both alloys are available in a range of wire diameters from 0.1 to 10 mm. The resistivities of the nichrome alloys and Kanthal at 20 °C are 108 and 135 μΩ cm, respectively, and remain relatively constant over their working temperature range (compared to tungsten which has a resistivity which varies from 5.5 μΩ cm at 20 °C to 85 μΩ cm at 2400 °C—see Fig. 3). However, at elevated temperatures in vacuum the Cr evaporates from the Ni/Cr alloy resulting in a decrease in resistance and corresponding rise in temperature. For this reason 800 °C is the maximum temperature at which this alloy should be used. Current temperature characteristics for Nichrome 80 (British Driver—Harris), are shown in Fig. 4.

The manufacturers of Kanthal (Kanthal) recommend that their wire should be preoxidized before use in vacuum. The alumina scale on the surface of Kanthal is more stable than the NiCr alloys and this, together with preoxidizing the wires, enables them to be used at slightly higher temperatures in vacuum.

A very significant factor in favor of the use of alloy resistance wires is their ability to be bent and shaped without breaking. Tungsten wire can present serious difficulties in this respect, but tantalum and molybdenum wires less so.

Perhaps the most user-friendly heating elements for use in the lower temperature range are Thermocoax heating elements marketed by Philips. These elements comprise a resistive core which is surrounded by a mineral insulant and the whole element is contained in a metallic sheath of inconel. Single and twin core heaters are available, and in the case of the twin core version the conductors come away from the sheath at only one end. Figure 5 gives plots of current versus temperature for a number of outer diameters of single core thermocoax heaters.

Temperatures above 1000 °C are achieved in resistively heated metal vapor sources using wires of one of the pure metals W, Ta, or Mo. In this respect their use is theoretically limited to 2560, 2400, and 1910 °C for W, Ta, and Mo, respectively, these being the temperatures at which their weight loss by evaporation does not exceed 1% in 100 h. However, their upper temperature of operation is in practice much lower than this due to their surface reactions at high temperatures, as discussed in Sec. IV C.

Based on the data of Jones and Langmuir,126,127 Spangenberg128 has compiled a very useful figure giving the lifetimes of tungsten filaments of different diameters for a range of filament currents; this is reproduced in part in Fig. 6; while it is recognized that filaments and heaters are used in different contexts, the data of Spangenberg128 provide figures on the upper limit of operation of tungsten resistance heaters. In Fig. 6 the lifetime is the time required for 20% of the mass of the filament to be evaporated. So, for example, a 0.5 mm tungsten wire operating at a current of 17 A has a lifetime of 1000 h. From the vapor pressure curves for tungsten125 this corresponds to a temperature of 2227 °C.

Taken with the figures for the radiation intensity and resistivity given in Table III, these data enable the performance of a resistively heated source using tungsten wire to be calculated.

Tungsten wire does not bend easily and, in addition to that, becomes very brittle when it has been raised to a high temperature. In contrast to tungsten, tantalum is not brittle either before or after heating. It has a maximum operating temperature close to that of tungsten and a similar value of resistivity and total emissivity. Hence, the data for tungsten wire in Table III can also be taken as an approximate guide to the upper limit of performance of tantalum wire.

Molybdenum wire is restricted in use to operating temperatures below 1900 °C. It is also a brittle material, although less so than tungsten. Of these three metals this is probably the least useful, while tantalum is by far the best and the most expensive.

The instantaneous surface power density for a wire of radius r and length l is given by

\[ \frac{1}{2} \pi r^2 \frac{\tau}{l} \text{ W/m}^2, \]
The most familiar of these is graphite which has a high negative coefficient of resistivity up to 600 °C after which it becomes positive. Graphite heater elements are machined from the solid and can operate at temperatures up to 2000 °C without serious reduction in working life due to evaporation.

Silicon carbide can be used as a heating element up to 1600 °C. This material, although brittle, can be machined after manufacture to produce heating elements of the required design. The main disadvantage of this material is the increase in resistivity produced by thermal cycling.

Molybdenum disilicate is another excellent heating element material and, although brittle at ambient temperatures, does not suffer from the aging process found in silicon carbide and can be used at temperatures up to 1700 °C.

One of us (B.S.) has recently used a Boralectric heating element in the design of a new oven for producing beams of Cr atoms. Boralectric heating elements are manufactured by Union Carbide and are available in a number of two- and three-dimensional shapes. They are made from pyroelectric boron nitride with an electrically conducting film of pyroelectric graphite as the heating element. It should be possible to design ovens for temperatures in excess of 1300 °C using these heater elements.

C. Ceramics

Ceramic materials form an essential part in the construction of vapor beam sources. Such materials are inorganic and require high temperature processing for their permanent shape and hardness.

By far the most important group of ceramics are the refractory oxides which are available as pure single refractory oxides, such as Al₂O₃, MgO, and ZrO₂, and complex refractory oxides such as 2BaO·SiO₂ and ZrO₂·SiO₂. Generally the single oxides are most readily available with alumina, magnesia, silica, and zirconia having the greatest application.

Ceramic bushes and washers can be manufactured to tolerances of ±1% whereas rods are typically manufactured to tolerances of ±5%. Higher tolerances are achievable by grinding, but these processes are found to weaken ceramics. Those ceramics which are high in alumina can tolerate a certain amount of grinding, but steatite and zircon are weakened by this process.

Similarly, cleaning ceramics with acids such as nitric acid which attacks the skin results in weakening, and also absorption of the acid in the pores of the material; generally air firing provides adequate cleaning.

Table IV lists a number of properties of the refractory metal oxides which are of interest to this paper, together with those of boron nitride; where more than one grade of ceramic is available, use of the highest purity material generally ensures maximum stability and resistance to chemical attack, where it might occur. Although zirconia has been included in Table IV, having a high melting point (2600 °C), it becomes electrically conducting at high temperatures and is therefore of limited use, particularly in connection with the radio frequency heating techniques discussed elsewhere in this paper. Although not a feature of Table IV it should be noted that the thermal conductivity of BeO is comparable with metals at
TABLE IV. Some properties of a number of ceramic materials.a

<table>
<thead>
<tr>
<th>Material</th>
<th>Melting point °C</th>
<th>dc resistance Ω cm</th>
<th>Coefficient of thermal expansion ×10⁻⁶</th>
<th>Thermal shock resistance</th>
<th>Temperature (°C) for vapor pressure of 10⁻⁵ mm Hg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>2050 830 °C 1095 °C 1875 °C</td>
<td>1×10⁸ 2.2×10⁷ 5.2×10⁷</td>
<td>25–800 °C 8.5</td>
<td>Good 1763</td>
<td></td>
</tr>
<tr>
<td>BeO</td>
<td>2520 630 °C 1100 °C</td>
<td>3.5×10⁸ 5.2×10⁷</td>
<td>25–800 °C 9.2</td>
<td>Very good 1748</td>
<td></td>
</tr>
<tr>
<td>ZrO₂</td>
<td>2720 340 °C 700 °C 2000 °C</td>
<td>2.1×10⁸ 2.3×10⁷ 0.59</td>
<td>0–1400 °C 5.0</td>
<td>Fair 2300</td>
<td></td>
</tr>
<tr>
<td>MgO</td>
<td>2800 800 °C 1600 °C</td>
<td>3×10⁶ 1×10⁶</td>
<td>0–1500 °C 16</td>
<td>Good 1227</td>
<td></td>
</tr>
<tr>
<td>ThO₂</td>
<td>3030 550 °C 970 °C</td>
<td>2.6×10⁷ 3.8×10⁷</td>
<td>25–800 °C 9.5</td>
<td>Fair 1971</td>
<td></td>
</tr>
<tr>
<td>BN b</td>
<td>&gt;3000</td>
<td>1×10¹⁵</td>
<td>Excellent</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

aData from Ref. 129.  
bBN data from Ref. 130.

By Corning. Like boron nitride, this material has no known toxic effects and machines well to high tolerances. It can be used in ultrahigh-vacuum environments.

A high thermal conductivity machinable ceramic called Shapal-M is supplied by Tokuyama. This material has a thermal conductivity of 100 W/m°C, some hundred times higher than that of Macor. Both Macor and Shapal-M have high mechanical strength.

V. MAIN TYPES OF ATOMIC BEAM SOURCES

A. Resistively heated atomic beam sources

Resistive heating has been used successfully in many cases for temperatures below 1200 °C (see, e.g., Kusch and Hughes, Lew, Pauly and Toennies, Hertel and Ross, Parr, Holland et al., and references therein), the crucible containing the sample being surrounded by a system of Mo or Ta wires kept in place by ceramic supports. Temperature control is achieved by regulating the current through the wires; noninductive wiring schemes are applied in order to minimize the magnetic fields created by the heating current. The hot core of the furnace is usually surrounded by a set of radiation shields which reduce the radiative heat losses. As an example, the atomic beam source developed by Hertel

Finally in this section, it is important to make specific reference to machinable ceramics as their availability is frequently essential in oven construction. In bulk form, PBN is only available in sheets. Much more useful to the subject of this paper is the HBC grade of diffusion bonded boron nitride. This is available in both sheet and bar form and, being relatively soft, machines easily.

Macor is a machinable glass ceramic which is supplied by Corning. Like boron nitride, this material has no known toxic effects and machines well to high tolerances. It can be used in ultrahigh-vacuum environments.

A high thermal conductivity machinable ceramic called Shapal-M is supplied by Tokuyama. This material has a thermal conductivity of 100 W/m°C, some hundred times higher than that of Macor. Both Macor and Shapal-M have high mechanical strength.

TABLE V. Maximum temperatures (°C) for surface-to-surface stability for some ceramics and refractory metals.a

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>W</th>
<th>Mo</th>
<th>Ta</th>
<th>ThO₂</th>
<th>ZrO₂</th>
<th>MgO</th>
<th>BeO</th>
</tr>
</thead>
<tbody>
<tr>
<td>BeO</td>
<td>2300 2000 1900 1600 2100 1900 1800</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MgO</td>
<td>1800 2000 1600 1600 2200 2000 1800</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ZrO₂</td>
<td>1600 1600 2200 1700 2200 2000 1900</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ThO₂</td>
<td>2000 2200 1900 2200 2200 2100</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>1300 1700 1800 1700 1700</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BN</td>
<td>1900 1500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>1400 1200 1000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

aData from Ref. 131.  
bObserved in this laboratory (K.J.R.)  
References 132.  
References 133.  
References 134.
FIG. 7. Schematic drawing of the vapour oven assembly developed by Hertel and Ross (Ref. 140).

and Ross is shown in Fig. 7. The oven is machined from stainless steel and fitted with separate upper and lower heating elements. Each heating element is contained in 12 open ended holes of 3.2 mm diam drilled in the oven wall; these holes are fitted with twin-bore ceramic tubes.

The heating wire (0.25-mm-diam Ta) is so arranged that in the upper oven section links between the adjacent ceramic tubes are made at the center of the oven while the links between the twin bores of any one ceramic tube are made at the top. In this way each tube contains a narrow hairpin producing a low-residual magnetic field. At the center of the oven the links between the tubes form a current ring. The lower heating element is formed in exactly the same manner, but again with the links between adjacent tubes being made at the center of the oven. This element produces a current ring directed opposite to the adjacent current ring of the upper heating element.

The oven aperture, which is of the corrugated metal-foil type described by King and Zacharias (see also Anderson et al.), is contained in a separate element which is screwed into the bulk of the oven. A close fitting mu-metal cover (0.4 mm thick) with an aperture for the vapor beam of 10 mm diam is placed over the whole of the oven (for information on magnetic shielding see, e.g., Firmenschrift Magnetische Abschirmungen, Telcon Metals and Mager and references therein). Thermocouple connections were made both at the base and the upper section of the oven.

At a distance of 25 mm above the aperture a current of 1.75 A (corresponding to 420 °C) flowing in both heater elements caused a magnetic field of less than 0.12 mG. This oven has been successfully used for the generation of alkali-metal beams and other elements requiring oven temperatures less than 1000 °C. The corrugated metal-foil multiple channel effuser has frequently been replaced by a bundle of hypodermic needles cut to length and compressed in the aperture. Alternatively a single aperture 1 mm diam and 10 mm long is used.

A more recent design by Ross while based on that of Hertel and Ross has enabled the crucible to be inserted and removed from the heater assembly, thereby enabling the sample to be recharged or changed without disturbing the heaters and thermocouples.

Deposition of metal on the oven aperture, especially on thin nozzles, can result in a drastic reduction of the atomic beam. Holland et al. used a cylindrical Mo crucible with a removable nozzle of 0.6 mm diam and 8 mm in length. The crucible was inserted in the cylindrical heating element formed by W wire noninductively wound on a machineable-glass former. Blocking of the furnace nozzle was avoided by increasing the groove density on the furnace glass by a factor of 2 on the upper half of the former.

In order to add flexibility, separate heating systems for the crucible and the nozzle have been developed. If crucible and nozzle are always operated at the correct temperatures the problems of clogging can be drastically reduced. The nozzle temperature should be at least 50 °C higher than the temperature of the crucible and the nozzle should be heated first in starting the source and cooled down last in shutting down the source.

In Fig. 8 the resistively heated atomic beam source used in atomic photoelectron spectroscopy by Becker and co-workers (Becker and references therein) is presented. The design is based on an earlier version described by Kobrin and co-workers. The main body of the furnace and the nozzle are made from Mo. The crucible is screwed to a long rod and
can be pressed against the outer oven wall, thus confining the metal vapor to the crucible and the nozzle. This design also allows the crucible to be removed for refilling without changing the alignment of the atomic beam source. It is clear that the prevention of any metal deposition in the nozzle is a prerequisite. This can be achieved by operating the nozzle at a higher temperature than the crucible. Commercial bifilar heating cables (Thermocoax, see Sec. IV B) are wound around the main body of the furnace and the nozzle. The cables are flexible and can easily be bent around the furnace components. The metal sheath and the insulating material protect the wires from aggressive vapors and gases and thus prevent, for example, troublesome embrittlement encountered for W wires operated at temperatures above 1200 °C. For temperatures below 900 °C these heating cables have replaced the open Ta or W wire heating systems.

The atomic beam source used by Hausmann\(^{152}\) and Hausmann et al.\(^{87}\) in their investigations of the photoelectron spectra of atomic Mg is schematically depicted in Fig. 9. The cylindrical crucible (8 mm i.d.) is heated by bifilar heating cables. Stainless steel cylinders insulated by ruby balls serve as radiation shields. The inner cylinder also holds the heating cables in place. The whole oven is surrounded by a water cooled Cu cylinder. The outermost Cu baffle is supported by this Cu cylinder. Hausmann et al.\(^{87}\) found that in order to prevent Mg crystals from growing on the baffles a hot, well-polished Mo baffle, in thermal contact with the crucible, had to be mounted between the crucible and the cold Cu baffle. At temperatures between 450 and 550 °C (25–35 W heating power) densities ranging from \(10^6\) to \(10^7\) atoms/cm\(^3\) have been achieved in the interaction region located approximately 25 mm above the crucible aperture. With a Mg charge of 14 g the vapor beam could be operated under stable conditions for up to 10 h.

A special furnace which can be mounted horizontally on the axis of a cylindrical mirror electron analyzer (CMA), has been developed by Sandner\(^{106}\) and is shown in Fig. 10. Modified versions of this basic model have been used with great success for the investigation of the x-ray ultraviolet (XUV) photoelectron spectra of ground state and laser excited alkali and alkaline earth atoms.\(^{78,45,153,157}\) The bifilar heating cable is wound around the cylindrical stainless-steel crucible. In the center of the crucible a tube (5 mm i.d.) protrudes from the back flange almost up to the front aperture. This tube permits the incoming XUV light to pass through the furnace onto a beam monitor mounted behind the furnace. The metal vapor is contained in the space between this tube and the outer wall of the crucible. The conical end section of the crucible directs the emanating atoms towards the interaction region. By this focusing of the atoms, target densities up to \(10^{13}\) atoms/cm\(^3\) have been achieved.

Krause and co-workers\(^{89,160}\) developed a resistively heated atomic beam source which is integrated in their system for electron spectrometry with synchrotron radiation\(^{161,162}\) (ESSR setup). A semischematic drawing is presented in Fig. 11. The cylindrical W or Ta crucible is surrounded by W or Ta heating wires embedded in Al\(_2\)O\(_3\) insulator tubes. The atomic beam, collimated by two baffles, propagates in the opposite direction to the photon beam which enters the electron source cell through a 2-mm-diam capillary. The energy and angular distribution of the  

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FIG. 9. Resistively heated atomic beam source developed by Hausmann (Ref. 157). 1 stainless steel crucible; 2 Cu baffle; 3 heating cables; 4 stainless steel radiation shields; 5 water-cooled Cu housing; 6 insulating ruby balls; 7 Ni–NiCr thermocouples.

FIG. 10. Schematic drawing of the atomic beam source operated on the axis of a cylindrical mirror electron energy analyser [Krummacher (Ref. 153), Gerard (Ref. 154), Bizau et al. (Ref. 45)].
FIG. 12. Schematic drawing of the resistively heated high temperature atomic beam source developed by Schmidt (Ref. 44) for the investigation of the photoion spectra of 3D metals.

FIG. 13. High temperature atomic beam source developed for photoelectron spectroscopy of atomic 3D metals (Refs. 163–167).

...trons emitted perpendicular to the photon beam are determined by three electron spectrometers (ESA). The maximum oven temperature is 1350 °C using a 0.25 mm tungsten heater wire.

Routine operation is possible at temperatures up to 1100 °C. The oven is typically operated at a temperature corresponding to a vapor pressure of $10^{-2}$ mm Hg, or slightly less. Although one oven charge lasts 30–60 h, deposition of metal on components generally requires cleaning of the source cell and the capillary after every 20–30 h of operation. This atomic beam source has been used with great success for the investigation of the electron spectra of atomic beryllium, cadmium, calcium, indium, lead, manganese, silver, and tin.

Temperatures up to 1800 °C have been reached with the resistance heated beam source developed by Schmidt for the investigation of the photoion spectra of three-dimensional (3D)-metal atoms. A schematic drawing of this source is presented in Fig. 12. The metals were contained in an Al$_2$O$_3$ crucible which was heated by radiation emitted from the W heating wires mounted around it. In order to reduce radiative heat losses a series of radiation shields surrounded the core of the oven and to protect the environment the whole oven was encapsulated in a water cooled Cu housing. Effective radiation shielding is essential for all high temperature sources.

A further problem in the production of metal vapor beams at all temperatures is that of the background gas of metal atoms. The level of seriousness of this problem does of course depend on the accommodation coefficient for the particular atoms and the surrounding surfaces. In the extreme case of an atom such as cadmium the whole of the system becomes contaminated within a short period of time. Attempts are made to minimize this problem by the use of liquid nitrogen cooled baffles and beam apertures, and for most atoms such baffles will prove effective.

The difficulties encountered in developing and operating high temperature atomic beam sources increase with temperature. For temperatures above 1200 °C Ta or W heating systems are very cumbersome to operate. Reactions with the insulating supports and the gases resulting from outgassing at the high temperature, together with the frequent cycling of the temperature required for refilling and cleaning, results in the wires becoming very brittle; after a few cycles they break very easily and have to be replaced.

A very effective method of heating first used by Gole and his collaborators is to use a resistively heated cylinder, usually graphite or tantalum, which surrounds the crucible holding the metal, to provide radiant heating. Because the heater is self-supporting this arrangement avoids the use of ceramic supports and their associated contact problems (see Table V).

The problem of the high magnetic fields which are associated with the high currents required with this heating technique was overcome by Sonntag and his collaborators in their investigations of the vacuum ultraviolet (VUV) photoelectron spectra of atomic 3D metals, undertaken at the electron storage ring DORIS in Hamburg. They developed a resistive heating system based on two concentric Ta cylinders. A schematic drawing of this atomic beam source is given in Fig. 13. The Al$_2$O$_3$ nozzle (inner diameter $\leq 2$ mm, length $\leq 20$ mm) and the Al$_2$O$_3$ crucible are surrounded by the two concentric Ta cylinders. At the top of the oven the cylinders are spot welded to a Nb ring and at the bottom to Nb conductors. The heating current, typically 100
A, runs upward in the inner cylinder and downward in the outer cylinder. The symmetry of this heating current in this system results in very low residual magnetic fields. The cylindrical heating elements also help to concentrate the heat on the axis of the system. A series of Ta radiation shields surrounding the heating elements reduces radiative heat losses, and a water cooled Cu housing covers the hot part of the source and protects the environment. All insulating parts are made from Al₂O₃ or machinable ceramics. The crucible rests on a high temperature thermocouple (Philips Thermo-coax high temperature W/Re thermocouple). The whole source is mounted on a NW 100 ultrahigh-vacuum flange and is inserted into the experimental chamber from below, the bellows allowing an alignment of the atomic beam. This source has been successfully operated in the temperature range from 800 to 1500 °C (power ≈ 2 kW).

A cylindrical heating element is also used in the Cr sublimation source developed by Roberts and Via. As shown in Fig. 14, a Cr rod is supported within a dual wall resistive heating cylinder. The latter is fabricated from pieces of 0.05-mm-(inner cylinder) and 0.12-mm-(outer cylinder)-thick Ta sheet which are spot welded together to form a continuous current path. Self-supporting cylinders made of Ta grid could be used as an alternative to the Ta sheet cylinders. The Cr vapor pressure reaches 10⁻² mm Hg at approximately 1350 °C, i.e., 500 °C below the melting point. Sublimation occurs from the entire surface area of the Cr rod, which is uniformly heated by radiation. As a result of being compact and well shielded the power requirements for this source are less than 750 W.

A magnetic-field free atomic beam source for angle and energy resolved measurements of photoelectron spin polarization at low kinetic energies has been developed by Schönhense. The basic idea of the beam source is that the reservoir containing the metal and the nozzle are heated by means of a stream of hot air. The hot air is produced in a mu-metal-shielded resistive heater a large distance (2 m) from the electron path. A scaled drawing of the air-heated vapor furnace is presented in Fig. 15. The nozzle (Ti 1 mm i.d.) and the stainless-steel tube connecting the nozzle and reservoir are double walled and hot air streams through the gap between the two coaxial tubes. The reservoir, which is Cu, with an inner stainless-steel crucible, has a double bottom; the hot air streams across the hollow between the two bottom plates. Temperatures up to 600 °C can be reached with this device.

B. Atomic beam sources heated by electron bombardment

Electron bombardment heating is common in thin film technology for materials requiring high temperatures for their vaporization. In electron bombardment evaporation sources the electrons are accelerated (typical acceleration voltages range between 2 and 10 kV) and directly focused on the evaporant by electric or magnetic fields (typical electron currents ≈ 100 mA).

However, there are three main problems that prevent the use of these evaporation sources for the generation of atomic beams: (i) the bombarding electrons in part pass through the vapor giving rise to excited and ionized species, (ii) scattered electrons can cause intolerable background counts, and (iii) the magnetic and electric fields are not well shielded and can render photoelectron spectroscopy, for example, impossible.

To overcome these difficulties a different approach has been taken by von Ehrenstein and by Wetzel and Prescher et al. The key to their success was their resorting to the heating of the crucible by a radiator which has been heated by electron bombardment. Figure 16 is a section of the source used by Prescher et al. The molten metal is contained in a tungsten, tantalum, or graphite crucible (1), the choice of the material depending on the sample. The atomic beam formed emanates through the orifice in the Cu cap (4) of the water cooled Cu cylinder (5), which surrounds the source. A lid (2) made from Nb, Mo, Ta, or C tightly seals the Cu cylinder. It is crucial that this lid prevents any electron created inside the cylinder from escaping into the outer region. The crucible (1) rests in a tubular extension of this lid. Four W filaments (6), separated by 90°, are mounted...
between a Nb ring (3) and an inner cylinder made out of a Ta sheet (8). An outer Ta-sheet cylinder (7) surrounds the filaments and the inner cylinder. Both cylinders are supported by electrically insulated stainless-steel rings (9,10). For heating the filaments, a current (7 V, \( \leq 60 \) A) is passed through the outer and the inner cylinder. Electrons emitted from the hot filaments are accelerated towards the tubular extension of the lid, which contains the crucible. A voltage of up to 1500 V can be applied between the filaments and the grounded lid and by this means the temperature of the crucible has been raised up to 1800 °C. The power required was less than 2.5 kW. Three outer stainless-steel rods (11) and one center rod (12) provide the electrical connections. Metal shields (13) protect the ceramic insulators (14) against metal contamination. The cylindrical geometry minimizes stray electrical and magnetic fields, which could otherwise adversely affect the performance of the electron energy analyzer.

This high temperature atomic beam source has been successfully used in the study of the VUV photoelectron spectra of atomic 3d metals,\(^{53,101}\) atomic rare earths,\(^{77,174}\) and K and Ca.\(^{76,175}\) An atomic beam source heated by electron bombardment based on the above system has also been used in the photoionization studies of the giant resonances of the lanthanides.\(^{176-178}\)

### C. Inductively heated sources

The production of high density atomic or molecular beams of systems whose vapor pressures only exceed 10\(^{-2}\) mm Hg at temperatures in excess of 2500 °C can only readily be achieved using inductive heating. However, even at temperatures as low as 1000 °C the method of induction heating is an attractive option; the cost of the radio-frequency power supply is probably the only factor which sets a lower temperature limit to the economic application of this heating technique, since the technique itself is arguably the most simple of the heating methods discussed in this paper. It is certainly the most direct method of heating, since it can be used to produce heating directly in the crucible containing the metal to be evaporated, the crucible holding the charge also acting as a susceptor.

Induction heating in the production of molecular beams was reported by Linevsky\(^{179}\) when a two stage oven employed rf induction heating for the second stage; Neubert and Zmbovi\(^{180}\) used a similar two stage arrangement, where the second stage was heated by rf induction in order to produce gaseous systems such as SmLi. The use of a single rf inductively heated oven to produce both atomic and molecular beams for photoelectron spectroscopic studies was pioneered and developed by Dyke and co-workers at Southampton;\(^{181}\) the technique has also been widely used by Ross and co-workers at Southampton\(^{172}\) in their studies of autoionization and Auger processes in metal vapor atoms. Schmidt\(^{182}\) has also used induction heating for photoionization studies of metal atoms and Aksela et al.\(^{75}\) reported using the technique for a study of the Auger spectrum of Au atoms.

The arrangement for induction heating in the present context is a crucible (which is also the susceptor) surrounded by a water-carrying work coil through which high frequency current, whose frequency ranges from 500 kHz to 1 MHz, is circulating. The rate of heating of the susceptor depends on two factors, the so-called skin depth and the thermal conductivity; the skin depth is given by

\[
\delta = \left( \frac{2\mu}{\rho \omega} \right)^{1/2} \text{mm},
\]

where \( \rho \) is the resistivity, \( \mu \) the permeability, and \( \omega \) the angular frequency of the work-coil current; the skin depth is the depth by which the induced current in the susceptor has fallen to \( 1/e \) of the value at the outer surface nearest the work coil. Eighty-six percent of the power is dissipated in the skin. The Joule heating produced in the susceptor is proportional to both the resistance or resistivity of the susceptor and the induced current. Hence there is a conflict in the choice of a suitable resistivity; high resistivity materials have lower induced currents. In practice a high resistivity material such as graphite is easy to heat. Tungsten susceptors, in contrast, require a high level of rf voltage before heating commences. However, the rise in resistivity with temperature means that heating becomes progressively easier as the temperature rises.

In order to minimize the contribution of heat flow to the heating effects of the susceptor the material should either be of a high thermal conductivity, or else the thickness of the wall of the susceptor should be approximately equal to the skin depth. The latter is a real possibility in a material such as graphite where the skin depth is approximately 1.8 mm at 1 MHz.

A typical arrangement which has been used by Ross and co-workers\(^{183}\) to produce metal vapor beams is shown in Fig. 17. The crucible acts as the susceptor and is supported on a ceramic tube. Heat loss by radiation is reduced by surround-
ing the crucible with a layer of carbon felt which is held in place by an insulating cylinder of either alumina or boron nitride; such an arrangement is suitable for temperatures up to 2000 °C. Although the aperture in Fig. 17 is shown screwed into the crucible, in many cases the aperture is made to be a slide fit into the top of the crucible in order to make its removal easier; an incorrectly balanced temperature distribution can result in the aperture being blocked and any system of threads can be seized-up with condensed metal.

Temperature measurements on this system are made by placing a thermocouple in contact with the base of the crucible; we have not found a convenient way to attach a second thermocouple to the top of the system. This means that the cooling of the aperture due to open space above it has to be overcome by varying the pitch of the work coil, and that achieving a suitable temperature distribution at the required operating temperature is a matter of trial and error (or experimentation).

The water-cooled work coil is typically made from 5-mm-diam copper tubing with 7 turns of inner diameter 24 mm being used to heat an oven 60 mm in length. The coil is usually wound as close to the outer radiation shield as possible, although, since the induced current in the susceptor has to equal the total current in the work coil (current x number of turns) this is not critical. Radiation losses, and therefore the heating up of other components in the system, are further reduced by surrounding the work coil with radiation shields; these shields should be slotted if they are close to the work coil in order to minimize induced currents.

The serious problem of maintaining a uniform temperature over the whole of the crucible, or even maintaining the aperture at a temperature some 50 °C in excess of the charge to avoid blocking, and in some circumstances dimer formation, is also encountered in reducing the crucible temperature from a high operating point. Blocking of the aperture can easily occur during this process if the power applied to the system is reduced too rapidly. We have found it necessary to reduce the temperature very slowly, usually by electronic means, until the temperature is equivalent to a charge vapor pressure of $10^{-4}$ mm Hg.

The highest temperature which has been achieved by this system is 1500 °C, although with the correct choice of materials it is capable of temperatures up to 2000 °C.

Figure 18 shows the arrangement typically used by the photoelectron spectroscopy group at Southampton for vaporizing samples requiring temperatures up to 2300 °C. The main difference between this and the arrangement in Fig. 17 is that the crucible is inverted to allow the vapor beam to be directed downward. In addition to that, a beam forming aperture is not used, but an inert carrier gas is employed to carry the source species into the interaction region. The crucible unscrews into two parts to allow loading. A very interesting feature of this system is the use of tungsten or molybdenum pinning in order to secure the oven to its ceramic mounting tube. Note also the use of an outer radiation shield, vital for such high temperature operation. The maximum temperature achieved with this system is 2300 °C using a graphite susceptor and crucible.

The inductively heated oven used by Schmidt is shown in Fig. 19. In contrast to Figs. 17 and 18, this system is highly engineered. The susceptor is molybdenum and it contains the ceramic crucible which contains the sample. The beam is crudely collimated by means of the diaphragm which is mounted on the top of the oven. The outer radiation shield is water cooled, and the induction coils are held in
MOLYBDENUM OVEN
WATER COOLED JACKET
TANTALUM OVEN SUPPORT

FIG. 19. Schematic drawing of the inductively heated source used by Schmidt and co-workers (Ref. 182).

place and insulated from this outer radiation shield by means of a ceramic tube.

When this system was used to study Al (vapor pressure 10\(^{-2}\) mm Hg at 1220 °C) by Malutzki et al.\(^{21}\) the susceptor, which also acted as the crucible, was made from tungsten. There have been no reports of temperatures higher than the above being employed with the system shown in Fig. 19, although in principle temperatures much in excess of 1220 °C should be possible.

Tungsten is used as a crucible for the production of atomic or molecular beams of species which require temperatures in excess of 2000 °C for their vaporization or which react aggressively with other elements. As has already been mentioned, at low temperatures (see Fig. 3) tungsten has a low resistivity and is difficult to start heating inductively. In addition to that, at temperatures above 1400 °C tungsten starts to form tungsten carbide when in contact with carbon wool (see Table IV) and, as shown in that table, reacts with most other ceramics above 1700 °C.

These problems have been overcome by Dyke and his co-workers\(^{184}\) by using as a susceptor a tantalum tube which surrounds the graphite oven and heats it radiatively. The arrangement is shown in Fig. 20 and is based on the work of Gule (see, e.g., Oldeboog et al.,\(^{185}\) Dubois & Gole,\(^{186}\) and Crumley et al.)\(^{62}\) who pioneered the use of graphite and tantalum cylinder radiators for heating crucibles; in the latter case the graphite and tantalum cylinders were heated by passing a high current through them. The tantalum radiator in Fig. 20 is in the form of a tube whose diameter is approximately 4 mm greater than that of the graphite crucible. Because of the surface-to-surface instability of tantalum with most ceramics above 1600 °C (see Table V) it is not possible to surround the tantalum with a radiation shield (such as carbon fiber) which makes contact with it, but a ceramic tube may be suspended between the tantalum and the work coil. Using this method temperatures of 2500 °C have been achieved.\(^{187}\) For these higher temperatures a water-cooled radiation shield was used outside the work coil.

When induction heating is used in conjunction with particle scattering or photoionization experiments it is impossible to screen the instrumentation from the rf signal applied to the work coil, and a way has to be found to make measurements while the rf signal is turned off.

In the case of the single phase generators used in the work of Bulgin et al.\(^{181}\) two of the four rectifying valves were removed from the rectifier bridge to produce a half-wave rectified wave form. The detecting electronics were then modulated in phase with the half-wave rectified signal to allow data acquisition only during the off part of the half-wave rectified wave form.

Much more satisfactory is to be able to modulate the rf supply itself. This was possible with a range of rf generators manufactured by Stanelco. A magnetically focused valve was used which could be modulated using an external 0–5 V pulse which corresponded to a power output from zero to maximum. The unit had rise and fall times of a fraction of a millisecond thereby enabling square wave modulation with variable mark-to-space ratio; square wave modulation results in more power per pulse being applied to the susceptor, and allows mark-to-space ratios of less than 1 to be used, thereby increasing data collection time; this system has been used by one of us (K.J.R.) for many years. It should be noted here that conventional rf generators can also be suitably modulated using a control unit which is available from Inductelec.

D. Loading reactive charges

There are both liquids and solids which might be required for atomic beam production and which react violently
In air. In the case of the very reactive elements, of which Cs is an excellent example, they are always supplied sealed in a glass ampule. Less reactive elements are usually sealed in an argon atmosphere. In opening and loading reactive samples one of us (K.I.R.) has replaced an argon glove box with an AtmosBag (Aldrich Chemicals) which is an inexpensive inflatable glove box. Inside the AtmosBag, reactive solids and powders are loaded into their crucible or oven and covered with a few mm of dry ether. In the case of materials such as Cs, the top is broken off the glass ampule and the sample covered by a few mm of dry ether. Once the material in the ampule is covered with dry ether the ampule is put into the crucible or oven and transferred to the vacuum system and the ether pumped off as the system is evacuated. For elements whose reactivity is not very high, flooding the crucible with argon gas followed by quick transfer to the vacuum system usually suffices to avoid oxidation.

The dangers of using open highly reactive elements cannot be understated; a halogen or CO₂ fire extinguisher should always be at hand.

As an alternative to handling reactive systems in the open laboratory, the ampules containing very reactive metals have to be cracked in the vacuum inside the furnace or be transferred from the cracking device to the crucible through a heated and evacuated transfer pipe. The first method was chosen by Prescher et al. in their study of Cs XUV photoelectron spectra. Spherical shaped glass ampules were broken by four metal pins which protruded from the lid into the stainless-steel crucible. By means of a bellows mounting the crucible could be raised with respect to the lid and thus the glass ampules could pressed against the pins and broken.

In the Cs source of Baum et al. the Cs ampule is cracked in a bellows assembly and the Cs is transferred through a heated filling pipe into their recirculating Cs oven. A cross section of this recirculating Cs oven is given in Fig. 21. Cesium vapor ascends by a linear feedthrough, to a 1-mm-diam orifice. A collimator with a diameter of 2 mm forms the beam. The temperature of the recycling funnel, including the surface of the collimator and recycling pipe, is kept just above the melting point of Cs. Thus the Cs which does not pass through the collimator condenses on these surfaces and flows down into the recycling pipe and from there back into the oven. The nozzle, the ascending pipe, and the oven itself can all be heated separately, allowing temperature settings which will prevent blockage. In order to reduce the dimer content of the atomic beam (see, e.g., Gingerrich, Sorokin and Lankard the nozzle is overheated by about 80 °C with respect to the oven. The collimator can be heated to clean the aperture. A stable beam with a density of $8 \times 10^8$ atoms/cm$^3$ at a distance of approximately 1.6 m from the nozzle has been achieved. The recirculation of the Cs saves material, prolongs the operating times, and drastically reduces the contamination of the environment. For their investigation of the superelastic scattering of spin-polarized electrons from sodium atoms, McClelland et al. used a recirculating sodium oven operating on the same basic principle.

Returning reactive materials to atmospheric pressure can be safely achieved by allowing them to cool to room temperature and then slowly admitting CO₂ to the system up to atmospheric pressure.

VI. MONITORING OF SOURCE PERFORMANCE

A. Temperature measurement

The determination of metal vapor source temperatures is essential both for the establishment and maintenance of a suitable operating regime; in the context of the present paper this means the monitoring of the temperature of the crucible containing the charge to be vaporized. For this purpose there is no doubt that the thermocouple is most suitable.

Temperature measurements below 1100 °C are most conveniently made with a chromel–alumel thermocouple. The chromel and alumel wires are strong, and the thermocouple has an electromotive force (EMF) of 45 meV at 1100 °C. Further, many digital instruments are available for chromel–alumel thermometers. Use of this thermocouple between 1100 °C and the theoretical limit of 1370 °C is not to be recommended; the thermocouple becomes unreliable and eventually fails when operating in the region of the upper limit of temperature.

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Difficulty is frequently encountered in ensuring good thermal contact between a thermocouple and the oven being monitored. This can be overcome by spot welding the thermocouple junction to a small tantalum disc, which in turn is lightly sprung against the oven.
For basic applications thermocouples can conveniently be made from lengths of cable of the required material, the junction being formed by welding the wires together; twisting the wires together is not satisfactory. Twin bore ceramic tubes are then used to carry the wires away from the junction.

An alternative method of construction is to use mineral insulated cable where the thermocouple cables are embedded in a closely compacted inert mineral powder and surrounded by a metal sheath which forms a hermetically sealed assembly. The sheath forms a useful protective cover, and can give the thermocouple sufficient stiffness to allow it to be pushed into contact with the point where the temperature measurement is required.

Omega Engineering and TC supply a range of thermocouple wires covering temperatures ranges up to 2200 °C, and in addition they offer mineral insulated thermocouples sheathed in stainless steel or nickel alloy. Of greater use for the high temperature vapor sources which operate at up to 2300 °C is the range of sheathed Thermocoax thermocouples offered by Philips. For temperatures up to 1100 °C they offer titanium, niobium, and molybdenum stabilized stainless steel sheaths, whereas for temperatures up to 2300 °C they offer tantalum, niobium, molybdenum, and rhenium sheaths.

A point of great importance in the context of using sheathed thermocouples in contact with graphite crucibles (graphite can be used for a wide range of source materials) is the surface reactivity of the materials used. As we discuss in the section on ceramics in this paper, BeO, an important insulator for sheathed thermocouples, only has surface to surface stability when in contact with Ta up to 1600 °C. Operation above this temperature means that reactions are underway which ultimately destroy the sheath. However, as is also shown in Table V, contact between the sheath and graphite results in reactions at temperatures as low as 1100 °C in the case of a Ta sheath; we overcome the direct corrosion of the Ta thermocouple sheath when it is in contact with a graphite oven by placing a protecting cap, made from Ta, over the thermocouple sheath.

For temperature measurements in the range of 1000-2200 °C there is no doubt that one of the range of tungsten–tungsten–rhenium or tungsten–rhenium–tungsten–rhenium thermocouples is most useful, having a range of electromotive force between approximately 9 and 22 meV over this temperature range. We have found the use of a pure-tungsten–tungsten–rhenium thermocouple very convenient; we have spot welded a thin tungsten–rhenium wire to the end of a tungsten rod, and in this way achieved precision location of the thermocouple junction. Perhaps even more useful is the range of sheathed tungsten-5% rhenium–tungsten-26% rhenium Thermocoax thermocouples mentioned above.

In addition to thermocouples, optical pyrometers may be used to monitor temperatures up to 2000 °C, but only to an accuracy of ±100 °C. There are a variety of instruments available ranging from hand-held systems to instruments where an optical fiber can be used to monitor sites which are impossible to access directly from outside the system. However, obtaining reliable data with these systems is not an easy matter.

An important aspect of being able to monitor source temperature is the ability to control the beam parameters described elsewhere in this paper. Temperature stabilization alone is easily achieved using a feedback circuit directly linked to the source power supply. A number of such systems are available commercially, although they always depend on thyristor control of an ac current source, and are therefore generally unsuited to the type of experiment which is the subject of this paper where ac fields are to be kept to a minimum.

B. Beam intensity

Collision experiments involving the use of an atomic beam are usually performed without an accurate knowledge of the atomic beam intensity, reliance being given to maintaining the temperature of the crucible containing evaporation source constant as a means of ensuring constant beam intensity. However, a number of methods do exist for both monitoring the beam stability and measuring beam intensities, and these are briefly discussed in this section.

For the range of atoms to which it can be applied, surface ionization is the most extensively used and most effective technique for monitoring atomic beams. This method is based on the observation by Langmuir and Kingdon that a hot tungsten wire produces 100% ionization of any Cs atoms which hit it; the resulting ion current is a direct measurement of the number of atoms striking the tungsten wire. In most applications of this technique there is no requirement to mass analyze the resulting ions.

The relationship between the number of neutral atoms leaving the hot wire $I$, and the number of atoms evaporating as ions $I^+$ involves the statistical weights of the ground and ion state of the atom together with the reflection coefficients for the particles from the surface, the so-called Langmuir–Saha equation. However, as a result of the uncertainties in these reflection coefficients and considerable uncertainties concerning the work function of polycrystalline tungsten wire, the equation which is generally used to describe the ration $I^+/I$ is

$$\frac{I^+}{I} = \exp \left( \frac{I - \Phi}{kT} \right) ,$$

where $I$ is the ionization potential of the incident atoms, $\Phi$ is the work function of the wire, and $k$ is Boltzmann’s constant. The work function of polycrystalline tungsten is usually taken as 4.5 eV and this means that for the alkalis Cs, Rb, and K the ionization efficiency is almost 100%. Hence for these elements this type of detector may be used for absolute beam intensity measurement.

The oxide of tungsten has a work function of approximately 6 eV and can be produced by heating the tungsten to a temperature of approximately 1200 °C in an oxygen pressure between $10^{-4}$ and $10^{-1}$ mm Hg. Such a filament can operate at temperatures up to 1300 °C without evaporating the oxide coating and can be used to monitor beams of atoms.
with ionization potentials up to approximately 6 eV. However, the ratio $I^+ / I_n$ given by the above equation may be in error.

The experimental arrangement for this type of detector is very simple. The hot tungsten wire is usually mounted on the axis of a cylindrical ion collector, which is in turn surrounded by a conducting shield. Atoms enter the system through slits cut in the side of the shield and collector. When this is used to monitor the profile of an atomic beam, arrangements are made to scan the system across the beam and to record the ion current as a function of position.

Electron impact ionization of atomic beams has long been used for beam monitoring and such detectors are frequently referred to as ionization gauge beam detectors since their design follows directly from that of the vacuum triode ionization gauge with the electron emitting filament mounted outside a cylindrical anode grid; the ion collector is a cylinder which surrounds both the anode and filament. The anode is operated at approximately +200 V, and the ion collector at −50 V. Atoms enter the detector through a small hole at the end of the gauge and in a direction along the axis of the cylindrical anode. The ion current is proportional to the beam intensity. However, absolute intensity measurements require a calibration to be carried out.

Ionization detectors find greatest application in molecular beam epitaxy (MBE) studies where the beam intensities are typically two orders of magnitude lower than those employed in collision studies. However, under these lower beam intensity conditions the beam ion current may become comparable with that of the residual gas. Schwarz has used an oscillating shutter to modulate the atomic beam and produce an alternating ion current which can be discriminated from the constant background gas signal.

Electron impact emission spectroscopy has also been used for atomic beam monitoring. A beam of electrons is used to excite the atomic beam and the intensity of one of the emission lines produced by the deexcitation is monitored. Again, this technique can monitor beam intensity as a function of time, but is unlikely to give absolute data on beam intensities.

Although monitoring mass deposition rates on a quartz crystal can be used as a method of determining beam intensities it has applications principally to MBE where deposition rates are low. However, these devices also require calibration if accuracy is to be obtained. Sauerbery and Lostis were the first to investigate this technique which uses a quartz piezoelectric crystal oscillator; the change in resonance frequency of the crystal is directly proportional to the change of mass resulting from the deposited atomic beam, and in this way deposition rates can be monitored. However, linearity is thought to be limited to a change of resonance frequency of 5% of the unloaded resonance frequency. Designs for crystal holders have been given by Behrnt and Love and Pulker. More recently Nyaiesh has published details of a voltage controlled oscillator circuit for use with quartz crystal oscillators and thin film thickness monitoring.

Crystal deposition rate monitors are available commercially. However, these systems are designed for thin film deposition control rather than high flux atom beam monitoring. A good example of this type of monitor is the Leybold Inficon system. Not only do they offer a crystal detector with a shutter assembly, but they also offer a detector head with automatic switching of six crystals to allow longer periods of operation. Such a system, although designed for thin film evaporation, may be appropriate for atom beam monitoring when used in conjunction with a shutter. Thus continuous monitoring would not be possible beyond a very limited period of time, but by using such a system intermittently it may be practical for atomic beam use.

Cold cathode discharge induced emission spectroscopy has been used by Sakai, Chen, Hiram, Murakami, and Ishida as a molecular beam flux monitor. The plasma-induced atomic emission is found to be linearly proportional to the source pressure and the log of the atomic emission inversely proportional to the source temperature. The sensor does not have a filament and can therefore operate over long periods of time without failure. So far use of this technique has only been reported for MBE studies.

Finally, some attention has to be given to optical techniques for beam monitoring. The wide availability of resonance sources means that resonance absorption can be used as a technique for monitoring beam intensities. While the sensitivity of this technique can be enhanced for molecular beams by reflecting the light beam back and forth through the molecular beam, the shorter wavelengths of the atomic resonance transitions generally preclude multipath crossings through the atomic beam. A further point to be considered in the context of absorption studies is the fact that the Beer-Lambert law of absorption does not always apply over wide ranges of concentration, and hence while resonance absorption is an excellent technique for beam monitoring, it is not to be relied upon for absolute intensity variation measurements.

Resonance fluorescence is several orders of magnitude more sensitive than resonance absorption. An arrangement for observing resonance fluorescence is discussed by Clyne and Monkhouse.

Laser induced fluorescence (LIF) is now a well-established technique for the investigation of atomic and molecular beam intensities. The technique was discussed in an early review by Kinsey. Some experimental detail of an
arrangement for observing LIF is given in Demtröder and more recently Kramer has described his experimental arrangement for studying LIF in considerable detail. The LIF technique is of particular importance when at high laser energy the fluorescence signal saturates and the fluorescence signal becomes proportional to the density of lower states.

Last in this context we should mention Rayleigh scattering. The scattering cross section for this process is very small for single atoms and increases rapidly with particle size; the technique is therefore well suited to the study of clusters, but has very low sensitivity in the study of single atoms.

Of the above techniques, the hot wire ionization technique is the easiest to use for absolute beam intensity measurement; the other techniques rely on calibration or experimental and theoretical data for the cross sections involved. However, to control the intensity of an atomic beam a signal from any one of the above monitoring techniques can be used to vary the output from the power supply determining the source temperature. Hence feedback loops can be incorporated into the experimental system, thereby maintaining a constant signal from the source monitor.

However, since source temperature is a direct control of vapor beam intensity it is generally easier to monitor the temperature with a thermocouple and use the thermocouple output as the control for the power supply controlling the heating process. In exceptional cases the evaporation surface area may change due to impurities, and under those circumstances the beam intensity will not be constant for a constant temperature. It is also found that certain MBE materials require a gradual rise in source temperature during evaporation in order to maintain constant beam intensities. However, in the context of collision experiments it is generally safe to assume that the maintenance of constant source temperature will ensure constant beam intensity, and in this context there is no doubt that the humble thermocouple is the most convenient and satisfactory means of maintaining constant densities in atomic beams.

As a further means of ensuring constant beam density in collision experiments it is usually possible to make periodic checks on the beam intensity by returning to an identifiable measuring point and checking for constancy in the observed signal. This self-consistency test is probably the best means there is of ensuring that atomic beam conditions are stable.