A new atomic beam source: The "candlestick"

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The design of a novel-type of atomic beam source which provides for long term, stable operation at high emission rates is reported. The heart of the design is the "candlestick" where liquid source material is transported by capillary action to a localized hot emission region. A surrounding cavity kept at the melting point for the source material shields the vacuum chamber walls from this region. The atomic beam escaping from the source is collimated, and uncollimated atoms are transported back to the liquid reservoir at the bottom of the "candlestick" by capillary action. This design has advantages over traditional oven designs: localized heating provides for large emission rates under high vacuum conditions, collimation is combined with recycling and conservation of source material, and the use of capillarity allows any orientation of the beam source. The source has been tested with sodium, and we believe that the design is useful for a broad range of applications including thin-film evaporation, molecular beam epitaxy, and semiconductor surface doping. With the low thermal mass of the emission section, the source could be optimized for pulsed mode operation. Furthermore, it is anticipated that the design ideas presented here could form the basis for a supersonic source with very high Mach numbers. © *1994 American Institute of Physics.*

I. INTRODUCTION

We report on a new type of atomic beam source developed in our laboratory to overcome a number of disadvantages of traditional designs. Our specific application is the generation of intense sodium atomic beams for use in laser cooling experiments in an UHV environment. However, we believe the ideas and design considerations involved can be used in other types of experiments and on other atomic systems, and that they have a broad range of applications such as in thin-film evaporation, molecular beam crystal growth, and semiconductor surface doping.

Our ideas have been stimulated by studies of heat pipes[1] and negative ion charge exchange canals of the type used in tandem Van de Graaff accelerators.[2] In particular, a major aspect of our design involves the use of capillary action to transport liquid metal from a cool reservoir to a localized hot evaporation region. Some of the evaporated atoms are permitted to escape the source as a collimated beam and the remainder are recycled to the reservoir through capillary or wicking action.

A schematic of the new design is shown in Fig. 1. The heart of the design is the candlestick consisting of a narrow metal cylinder filled with wire cloth which acts as a wick. Heating is provided by a small filament resistive heater placed close to the emission hole at the top of the candlestick. The heater and source regions are mechanically isolated to protect the filament from contamination by source material. The candlestick is mounted in a cylindrical copper cavity lined with a few layers of wire cloth which returns uncollimated atoms to the reservoir. The lining is in physical contact with the wire cloth that serves as the wick in the candlestick. Sodium is loaded into the copper chamber from the top by removing the chamber's lid. After pumpdown the chamber is brought to a temperature just above the melting point of sodium. Capillary action causes sodium to rise to the hot region around the emission hole where the emission rate is controlled by a vapor pressure corresponding to the local elevated temperature. The copper cavity contains a collimation hole from which the atoms emitted from the source escape. The collimation of the atomic beam is determined by the sizes of the emission and collimation holes. The wicking action of the cloth lining the cavity wall keeps the collimation hole from clogging up—a well-known problem with simpler designs.

The copper chamber is mechanically connected to and is supported by a solid 1/4 in. copper vacuum feedthrough which also serves to thermally couple the chamber to external heating and cooling. The entire assembly is compact and mounts in a standard 2 3/4 in. flanged UHV four-way cross. The flange[3] we have chosen with the above-mentioned copper feedthrough also provides feedthroughs for two thermocouple pairs as well as for the heater filament current.

The heater element is shown schematically at the top of the candlestick in Fig. 1. It was wound with 18 turns of 0.25 mm diam tungsten wire around a 1/8 in. diam by 1 in. long alumina insulator. A 0–80 threaded molybdenum rod attached to the removable cap of the candlestick passes through the alumina insulator which is captured with a nut from below. The filament is electrically grounded to the nut by a spot weld. The cap and filament assembly are easily removed from the candlestick for repair or replacement.

Our design has several significant advantages over conventional ovens. To obtain high emission rates, only a very localized part of the system is heated and this hot region is shielded by the copper housing which is kept at a much lower temperature. This solves the common problem of vacuum chamber wall outgassing with resulting high back-

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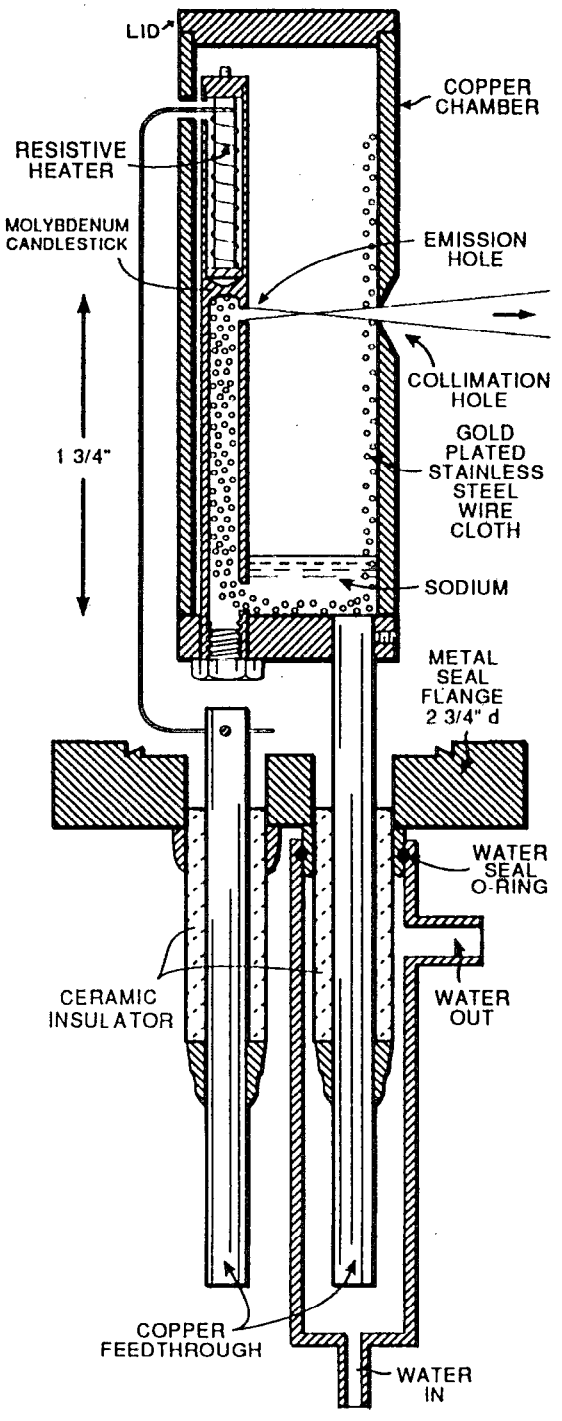

FIG. 1. Cross-sectional view of the candlestick atomic beam source. The assembly is mounted on one flange and fits inside a 2 3/4 in. UHV cross.

ground pressure. Furthermore, the source collimation prevents contamination of chamber walls with source material. Finally, the need to open the system for source recharging is greatly reduced due to the recycling feature of the design.

Localized heating through a candlestick, low power requirement, ease in reloading, operation with sodium, the fact that the source can be placed at any angle (including the position where the exit hole points downward), and the possibility for very high pumping speeds obtainable in the copper chamber, as described below, represent advances over the cesium source presented in Ref. 4.

Section II contains a detailed outline of design and fabrication considerations, followed by Sec. III which presents performance data in terms of emission rates as a function of temperature.

II. DESIGN

The first requirement on the wire cloth material is that liquid sodium must wet it at low temperatures. Inspired by studies of heat pipes we have chosen to use 304 stainless-steel wire cloth.[1] From these studies it is also apparent that a sealed bakeout at 800–900 °C for several days in the presence of sodium is necessary to initiate the wicking process. Otherwise the wetting process is prevented by surface oxides. For a beam source, which is an open system, such a treatment would be impossible since all the sodium would escape. However, we have found a trick which allows for low-temperature wicking activation: gold plating. Our experiments have shown that electroplating stainless steel with a thin layer of gold results in wetting at the melting point of sodium without extensive cleaning or heating procedures. Stainless-steel wire cloth is degreased and commercially electroplated with a 0.1 μm gold layer and with a thin intermediate layer of nickel (nickel strike) to provide for adhesion of gold. Our conjecture is that the stainless-steel surface is electropolished as part of the plating process and that the coverage with gold seals the clean surface. Liquid sodium wets the inert gold surface and—as seen from phase diagrams for the gold-sodium system[5]—it actually dissolves the gold layer and leaves behind a clean surface of stainless steel (or nickel) on which wetting takes place. Much thinner layers of gold could presumably be used—the basic requirement is that the whole surface should be covered. After the wire cloth is wetted, the system is very robust to openings for repairs and reloading, as long as it is kept under an argon atmosphere.

The initially wetted wire cloth released hydrocarbons when heated. We assume their origin to be trapped pockets of plating solution in the gold layer. A residual gas analyzer showed the released hydrocarbons to consist mainly of CH_3 and CH_4 molecules.

The presence of gold dissolved in the sodium can cause the slow formation of sodium-gold alloys at the emission hole. This process is seriously accelerated when the amount of remaining sodium is small. As seen from the phase diagrams in Ref. 5, formation of gold alloys with high melting points takes place at high gold concentrations. In the worst case, these alloys will clog up the emission hole and will have to be removed, which is easily done. However, not allowing the sodium level in the reservoir to drop too low (to less than $\approx$1 g) alleviates this problem.

The candlestick was machined of molybdenum with an outer diameter of 1/4 in. and a wall thickness of 0.050 in. in the source region, and 0.025 in. in the filament section. Stainless steel at high temperature releases large amounts of hydrogen so molybdenum was chosen to minimize the background pressure.

There are several requirements which will set the ultimate design parameters. Some specific choices here are influenced by our need for a sodium source. To keep the emission point hot and the reservoir cool, the thermal resistance of the candlestick has to sustain a temperature gradient of several hundreds °C for a heater power input (tens of watts) small enough to be dissipated in a reasonably simple way. This sets a lower limit for the length of the candlestick. The upper limit is set by the height of capillary elevation of sodium obtainable in the wick. We picture the wick as an array of tubes formed by adjacent layers of wire cloth and with diameters of the order of the wire size. We used cloth with 250 openings per inch and a wire diameter of 0.0016 in.

The capillary height h is given as the height at which gravity balances the capillary force, which is proportional to the surface tension, T (175 dyne/cm for sodium), and inversely proportional to the radius of curvature, R, of the surface,[6] yielding

$$h=\frac{2T}{Rg\rho}. \tag{1}$$

For zero wetting angle and small tube diameter the radius of curvature equals the radius of the tube. Furthermore, g is the gravitational acceleration and ρ the mass density of liquid sodium (0.9 g/cc). This results in a maximum capillary elevation of 95 cm.

Using the geometry of the molybdenum, sodium, and stainless-steel components and their thermal conductivities, we calculate that a candlestick length of 4.5 cm is enough to sustain a temperature gradient of 250 °C with a power input of 23 W. Note that this length is significantly lower than the capillary height, which will be of importance for the delivery rate considered next.

In order for the emission rate to be controlled by the vapor pressure (1×10^{-4} atm at 350 °C) at the emission point, the delivery rate must be large enough to balance the maximum emission rate envisioned with the system. The delivery rate is calculated by modeling the capillary transport of sodium as a viscous flow in tubes of the given diameter. The calculation requires the average velocity $\bar{v}$ of sodium in the capillary, which is proportional to the pressure gradient, ΔP, across the tube[6]

$$\bar{v}=\Delta P\cdot\frac{a^2}{8\mu l}. \tag{2}$$

Here μ is the viscosity of sodium, l the length, and a the radius of the capillary tube. The pressure gradient is given as the difference between the capillary and gravitational force per unit cross-sectional area of the tube

$$\Delta P=\frac{2T}{R}-g\rho l. \tag{3}$$

The overall delivery rate is given by the product of the average velocity, the density of sodium, and the cross sectional area of the candlestick which in our case is much larger than the area of the capillary tubes. For a candlestick length of 4.5 cm, the delivery rate is on the order of 10^{22} atoms per second which, as seen below, is four orders of magnitude larger than emission rates in the temperature range from 350 to 400 °C.

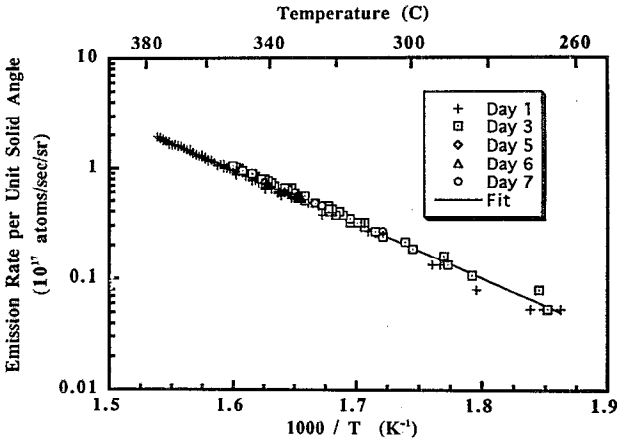

FIG. 2. Sodium emission rate per unit solid angle as a function of temperature. The different sets of data points correspond to different series of measurements and show the stability of the source. Note the exponential dependence on inverse temperature.

This large delivery rate allows the possibility for source operation in a supersonic regime at higher temperatures.

III. EXPERIMENTAL DATA

In this section performance data for the source are presented. Sodium emission rates per unit solid angle as a function of temperature are shown in Fig. 2. The rates were obtained by using a quartz crystal thickness monitor.[7]

An exponential dependence of emission rate on inverse temperature is clearly seen in the figure where the data points on the semilog plot fall on a straight line. From the slope of the line we determine the latent heat of vaporization at a constant temperature for sodium. The value obtained is 94 kJ/mole in the temperature range from 260 to 380 °C. In Ref. 8 the heat of vaporization at the boiling point of 883 °C for a pressure of 1 atm is tabulated as 98.01 kJ/mole.

The source performance shows high stability and reproducibility: emission rates were constant for weeks of continuous running and the same performance curve was obtained in several runs. Furthermore, emission rates are insensitive to the temperature of the copper cavity—the same rates were obtained for cavity temperatures in the range from 130 to 150 °C. The measured rate per unit solid angle for an emission point temperature of 350 °C corresponds to a total emission from the candlestick of 10^{18} sodium atoms per second which agrees with calculations based on a Boltzmann velocity distribution and a vapor pressure corresponding to that temperature. The expected collimation of the emitted atomic beam was confirmed by optical fluorescence.

The following procedure was used to obtain the emission curves shown. The source was assembled with one thermocouple attached to the back of the candlestick at the height of the emission point and another to the bottom of the copper cavity. The system was pumped down and baked for 1 1/2 h with the chamber walls and the copper chamber at 150–170 °C and the candlestick at 400 °C. The copper cavity was then loaded in an argon atmosphere with 7.4 g of sodium through a 2 3/4-in.-flanged loading port placed above the source. The system was pumped down and the copper cham-

TABLE I. A list of examples of potential source materials for the candlestick atomic beam source. The list has been constructed by considering melting point, vapor pressure (Ref. 9), surface tension (Ref. 8), viscosity (Ref. 10), and heating power requirement for each element. Furthermore, we have taken into account materials compatibility in suggesting materials for wire cloth, candlestick, and cavity (Refs. 11, 12, and 5) (stainless-steel 304 is abbreviated to s.s.). The suggested wire cloth materials are all commercially available. It should be stressed that except for sodium we have not performed experiments on the candlestick source with these materials and that preliminary wetting experiments are necessary in each case. In the table, we only list materials with melting points lower than 660 °C in order to minimize radiation from the cavity to the surrounding vacuum chamber walls. (For materials with higher melting points, a surrounding, cooled shield is necessary.) For each source material we show the melting temperature, T_{melt}, and the temperature, T_{emis}, at the emission point corresponding to a vapor pressure of 10^{-4} atm (except for antimony for which a vapor pressure of 10^{-2} atm was used). With an emission hole diameter of 2 mm and a collimation of 1/25 sr (as in the design shown in Fig. 1) a vapor pressure of 10^{-4} atm would correspond to a growth rate of a few monolayers per second over an area 2.3 cm in diameter on a substrate at a distance of 11 cm from the source.

Lithium T_{melt}=453 K T_{emis}=900 K Wire cloth: S.s., Ta, Mo, or W Candlestick: Mo Cavity: S.s.	Sodium T_{melt}=371 K T_{emis}=625 K Wire cloth: S.s. (gold plated) Candlestick: Mo Cavity: Cu	Potasium T_{melt}=337 K T_{emis}=550 K Wire cloth: S.s., Mo, Ta, or W Candlestick: Mo Cavity: Cu or s.s.	Rubidium T_{melt}=312 K T_{emis}=500 K Wire cloth: S.s. or Mo Candlestick: S.s. Cavity: S.s.	Cesium T_{melt}=302 K T_{emis}=500 K Wire cloth: S.s. Candlestick: S.s. Cavity: Cu
Aluminum T_{melt}=933 K T_{emis}=1700 K Wire cloth: Graphite fibers Candlestick: Graphite Cavity: Graphite	Gallium T_{melt}=303 K T_{emis}=1500 K Wire cloth: W or graphite fibers Candlestick: Graphite Cavity: Graphite	Indium T_{melt}=430 K T_{emis}=1300 K Wire cloth: W or Mo Candlestick: Mo Cavity: S.s. or Mo	Thallium T_{melt}=577 K T_{emis}=1000 K Wire cloth: W or Ta Candlestick: Ta Cavity: S.s. or Ta	Tin T_{melt}=505 K T_{emis}=1700 K Wire cloth: Mo Candlestick: Mo Cavity: Mo or Al_2O_3
	Lead T_{melt}=601 K T_{emis}=1100 K Wire cloth: W, Mo, or Ta Candlestick: Mo Cavity: Mo or Al_2O_3	Antimony T_{melt}=904 K T_{emis}=1300 K Wire cloth: Graphite fibers Candlestick: Graphite Cavity: Graphite	Bismuth T_{melt}=545 K T_{emis}=1100 K Wire cloth: Ta, W, Mo, or graphite Candlestick: Mo or graphite Cavity: Mo or graphite	

ber was heated from the outside through the copper feedthrough. The temperature of the cavity was kept at 110–150 °C for 12 h to allow for melting, wetting, and cleanout. Finally, the filament was heated and the input power was dissipated by water cooling the copper feedthrough from the outside. With 23 W of input power, the emission point was kept at a temperature of 340 °C and the copper cavity at 150 °C.

The system subsequently ran for 180 h, corresponding to an emission of 26 g of sodium through the hole in the candlestick. This shows that the recycling process is working: every remaining atom has been around the system three or four times. The accumulated hours were obtained partly by turning the system on for shorter periods of time and partly by running continuously for a week.

To put the effect of recycling into perspective it should be noted that the present load of sodium is projected to last for 7500 h of continuous running at an emission rate of 10^{18} atoms per second from the candlestick and with a solid angle of collimation of 1/25 sr.

Although we have only utilized the candlestick source with sodium, we anticipate that extension to other liquids will be straightforward. In Table I we give a list of examples of potential source materials. Thermal conditions and choice of materials for wire cloth, candlestick, and cavity must be changed to match the specific source material chosen. Melting points and emission point temperatures are given for the elements in the table along with materials suggestions. Except for sodium, we have not tested the source with the elements listed, and in each case preliminary wetting experiments will be necessary to assure proper wicking action.

IV. DISCUSSION

We have designed and tested a new atomic beam source which provides for large emission rates under high vacuum conditions. Uncollimated atoms are recycled which enhances the time period between successive reloads by orders of magnitude. The basic requirement to the functioning of the device is wicking action. Gold coating the stainless-steel wire cloth allows for low-temperature wetting activation for the sodium-stainless-steel system. The versatile design has potential use in many applications such as thin-film evaporation, molecular beam epitaxy, and semiconductor surface doping. Another important feature is the low thermal mass of the emission system and we envision that it could be optimized for pulsed mode operation.

A slice in the copper cylinder behind the candlestick could be introduced without leakage of source material resulting in high pumping speed and low pressures in the copper chamber. The simultaneous utilization of condensation and recycling would make the design a potential basis for a supersonic source with high Mach numbers.

ACKNOWLEDGMENT

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[1] G. M. Grover, T. P. Cotter, and G. F. Erickson, J. Appl. Phys. **35**, 1990 (1964); see also P. D. Dunn and D. A. Reay, *Heat Pipes*, 4th ed. (Pergamon, New York, 1994).
[2] M. Bacal and W. Reichelt, Rev. Sci. Instrum. **45**, 769 (1974).
[3] Insulator Seal Incorporated Part No. 9392031.
[4] R. D. Swenumson and U. Even, Rev. Sci. Instrum. **52**, 559 (1981).
[5] *Binary Alloy Phase Diagrams*, edited by T. B. Massalski (American Society for Metals, Metals Park, OH, 1986).
[6] A. Sommerfeld, *Mechanics of Deformable Bodies* (Academic, New York, 1950).
[7] Sycon Instruments STM-100 thickness monitor.
[8] *American Institute of Physics Handbook*, 3rd ed., edited by D. E. Gray (McGraw-Hill, New York, 1972).
[9] *Selected Values of the Thermodynamic Properties of the Elements*, edited by R. Hultgren *et al.* (American Society for Metals, Metals Park, OH, 1973).
[10] *Handbook of the Physicochemical Properties of the Elements*, edited by G. V. Samsonov (Plenum, New York, 1968).
[11] *Landolt–Börnstein Zahlenwerte und Funktionen aus Physik, Chemie, Astronomie, Geophysik und Technik*, 6th ed., edited by H. Borchers *et al.* (Springer, Berlin, 1965), Vol. 4, Pt. 2.
[12] *Thin Film Evaporation Source Reference* (Mathis, Long Beach, CA, 1987).