Appl. Phys. B 35, 65–69 (1984)

Applied Physics B Photophysics and Laser Chemistry

A Superheated Na Cell for X-Ray Photoionization Experiments

P. J. K. Wisoff and R. G. Caro

Edward L. Ginzton Laboratory, Stanford University, Stanford, CA 94305, USA

Received 22 May 1984/Accepted 6 June 1984

Abstract. The performance of a superheated alkali vapor cell, with a geometry suitable for x-ray photoionization experiments, is described. An internal heater was used to superheat sodium vapor in a split-wick heat pipe. At a Na pressure of 20 Torr, the transmission through the cell in the region of the $C-X$ molecular band of Na_2 increased to 50% from the 5% observed when the cell was operated in a conventional heat-pipe mode. In the presence of a Na atom density of $2\times10^{17}\,cm^{-3}$, the average molecule density over the length of the cell was measured to be $2.5\times10^{15}\,cm^{-3}$. In the hot central region of the superheated cell, the molecule density was predicted to have been reduced by a factor of 50 to $3\times10^{14}\,cm^{-3}$.

PACS: 35.80, 07.20K, 07.85

Recently, two new methods have been developed which make use of x-rays, emitted from a laser-produced plasma, to excite large populations in energetic metastable levels of atoms [1] and ions [2, 3]. Levels of this type serve as population "storage" states in a number of three- and four-level xuv laser systems that have been proposed in the literature [4–6]. Many of these systems involve alkali-metal atoms or ions as the lasant species [4–6], and their investigation thus requires the use of an appropriate alkali-metal vapor cell.

The production of excited alkali atoms and ions by these x-ray excitation techniques occurs most efficiently at relatively high alkali densities (10^{17}–$10^{18}\,cm^{-3}$). At these densities, significant alkali dimer populations (1–10%) are present in a saturated vapor [7]. The strong absorption bands of these dimers make the measurement of excited-state populations by absorption spectroscopy [1–3] difficult, if not impossible. In addition, the eventual realization of xuv laser action in one of the systems under discussion would require the rapid transfer of population from a metastable "storage" state to the upper laser level by an intense visible laser pulse. Molecular absorption would limit the wavelength regions in which such a transfer laser could be used. Finally, the excitation techniques that have recently been demonstrated utilize the x-rays from a laser-produced plasma to excite the surrounding atomic vapor. This plasma is created when an intense laser beam is focused onto a solid metal target which is placed inside the alkali vapor cell. The passage of the laser beam through the vapor may be affected by the strong molecular absorptions.

For these reasons, as well as for more general applications in fields such as nonlinear optics, it was desirable to develop an alkali vapor cell that was free of alkali dimer molecules. In this paper, we describe the operation of a relatively molecule-free, superheated Na vapor cell which has a geometry compatible with the use of x-ray photoionization excitation techniques. This cell is capable of operating at Na atom densities of at least $2\times10^{17}\,cm^{-3}$. The average Na_2 population over the length of the cell has been measured to be reduced from the saturated-vapor dimer density to $2.5\times10^{15}\,cm^{-3}$. This resulted in an increase of the average transmission in the region of the $C-X$ band (3600–3200 Å) from 5% to 50%. The cell should also prove satisfactory for producing large densities of relatively molecule-free atomic vapors of the heavier alkalis K, Rb, and Cs.

Figure 1 shows the dependence on temperature of the fraction of alkali metal which is present as dimers in a vapor at a constant pressure of 20 Torr [7]. As the

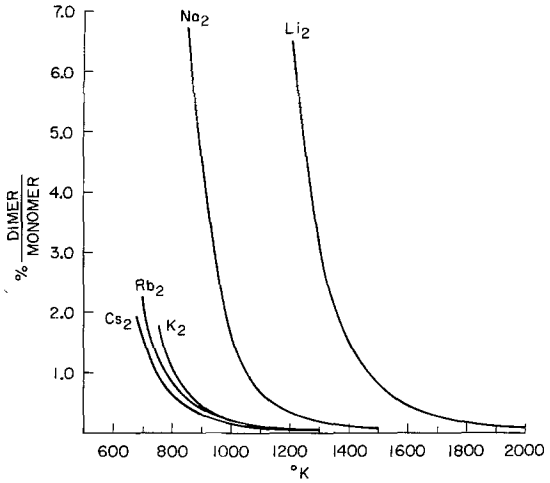

Fig. 1. The percentage of alkali-vapor molecules as a function of temperature, for superheated vapor at 20 Torr of pressure

vapor is heated above the temperature at which the saturated pressure is 20 Torr, thermal dissociation of the molecules progressively occurs. A reduction of the density of alkali molecules can thus be achieved by superheating the vapor to the point, where adequate thermal dissociation of the molecules has taken place. Wyatt and Cotter [8] have used two different methods to construct superheated Cs vapor cells in which reduced fractional densities of molecules were produced. One approach was to use a heat pipe with a split wick [9] in which the central portion of the vapor was superheated with external heaters. The alternative method was to suspend a supplementary heating coil inside the heat pipe in order to superheat the region of vapor along the axis of the cell. The former approach has been subsequently extended by Harris et al. to an operating temperature of 1200 °C [10]. The split-wick design, however, requires careful adjustment to minimize the effect of molecular absorption in the heat-pipe transition zones and to avoid the formation of unstable and turbulent interfaces that block transmission through the cell. For these reasons, and because it appeared better suited to the complicated cell geometry required for x-ray photoionization experiments, we constructed a cell which had a split-wick design but which was superheated by an internal heating element. As is evident from Fig. 1, the temperatures required to superheat Na vapor are considerably higher than for Cs vapor, and thus the design of the internal heating element required the solution of a number of technical problems. Since this work represents the first report of the operation of a superheated cell containing an alkali other than Cs, the performance and construction of the cell are described in detail.

1. Cell Design

The superheated cell is illustrated in Fig. 2. The inset shows a simplified picture of the whole cell. The geometry is designed to be suitable for laser-plasma x-ray photoionization experiments in which a 1.06 μm laser beam is focused onto a target which is inserted through one arm of the cell [1–3] X-rays, emitted from the laser-produced plasma, propagate into the surrounding alkali vapor producing hot

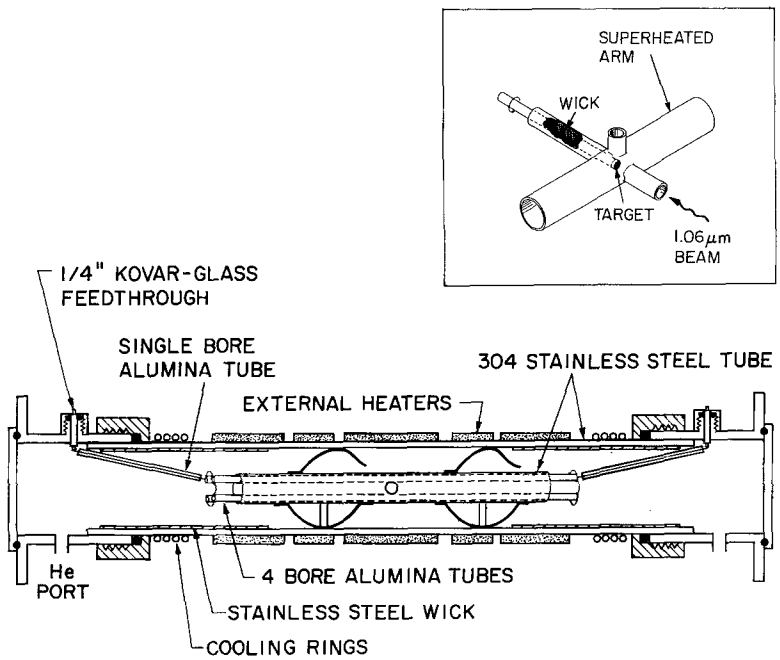

Fig. 2. The central heating element is shown inside the superheated arm of the alkali-vapor cell. A simplified schematic of the whole cell is shown in the inset

photoelectrons which subsequently excite the alkali atoms. The difference between this cell and that used in previous work [3] is that, in this case, one arm of the cell was superheated. This allows the "probe" or "transfer" laser beams to pass through the photoionized vapor without being affected by molecular absorption. The detailed construction of this superheated arm is shown in Fig. 2.

The principal technical problems associated with the construction of the cell were the maintenance of structural rigidity in the internal superheated section of the cell and the use of insulating materials and heater elements compatible with Na vapor at temperatures up to 1300 K. In addition, it was necessary to prevent the formation of liquid Na droplets on the heater wire as these could cause a short circuit between the heater wire and the cell wall and the consequent destruction of the heater. Structural support of the heaters was provided by a type 304 stainless steel tube of 0.75″ diameter that was supported inside the 2″ diameter cell arm by four stainless steel spring supports. Holes were cut in this tube for insertion of the target and to allow circulation of the Na vapor. Four alumina (99.8% purity) rods were strapped to the inside of this tube, each of diameter 0.187″, 76 cm in length, and containing four longitudinal holes of 0.047″ diameter. Tantalum heater wire (0.025″ diameter) was threaded through these holes to give a total length of wire of 12 m. The heater wire was connected to a power supply by vacuum feedthroughs. It proved necessary to sheath the connections from the heating element to the feedthroughs with an alumina tube to prevent liquid Na from condensing on these connections. In the superheated mode of operation, 850 W of power was applied to the inner heating element.

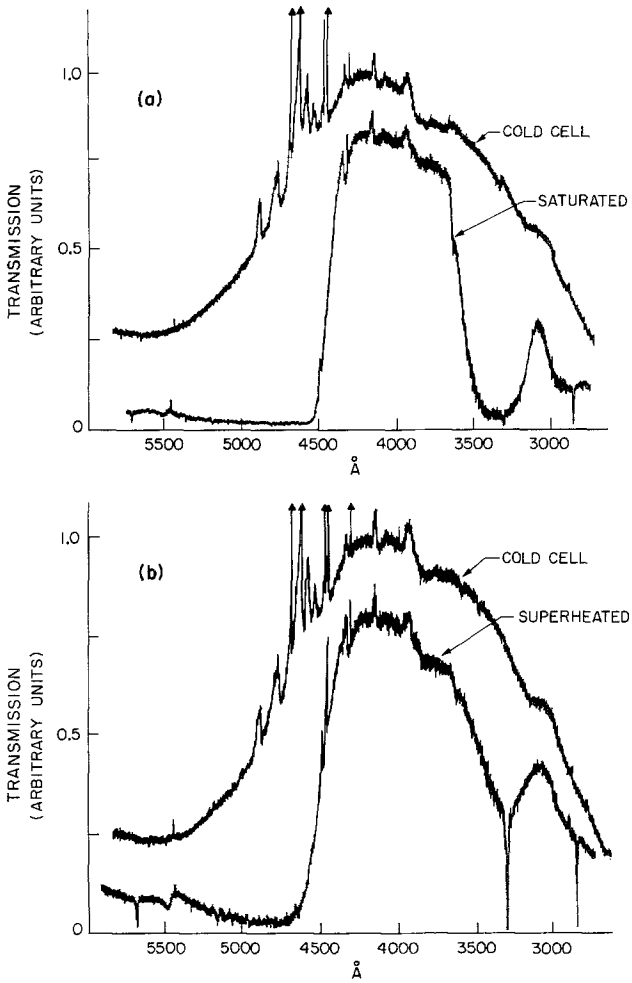

Fig. 3a and b. Xe arc lamp transmission scans: (a) saturated-vapor mode; (b) superheated-vapor mode

2. Cell Performance

By appropriate adjustment of the heaters, the cell could be operated in two modes. With the inner heater turned off, the cell operated as a conventional heat pipe. Under these conditions, the entire vapor column was in equilibrium with the liquid metal on the wicks at each end of the cell arm. In the second mode of operation, the cell arm was superheated. The internal heater alone provided heat to the 2″ diameter arm while the connecting 1″ diameter arms were heated externally. The central superheated section was held at a temperature of 1300 K. In both cases, the wicks remained at a temperature of 865 K defined by operation of the cell in a heat-pipe mode at 20 Torr of Na with 20 Torr of He buffer gas.

The performance of the superheated cell was monitored by observing the transmission through the cell of light from a Xe arc lamp. The spectrometer resolution was approximately 5 Å. The results of this experiment are shown in Fig. 3. The traces labeled "cold cell" represent the light transmitted when the cell was at room temperature. These traces record the lamp spectrum and the response of the detection system. Also shown in Fig. 3a is the absorption of the cell when operated in the conventional heat-pipe mode at a saturated vapor pressure of 20 Torr. Large regions of very low transmission due to molecular absorption are evident, particularly in the $B-X$ (5040–4560 Å), $C-X$ (3600–3200 Å), and $D-X$ (3325–3030 Å) bands [11]. In the region of the $C-X$ band, the molecular absorption cross section has been measured [12] to be approximately $4\times10^{-18}\,\mathrm{cm}^2$ at 3500 Å. This cross section was measured with an apparatus of resolution $6\,\mathrm{cm}^{-1}$. This is comparable with the resolution of the apparatus used to obtain the data of Fig. 3. Using the data of Fig. 3a, and the measured vapor column length

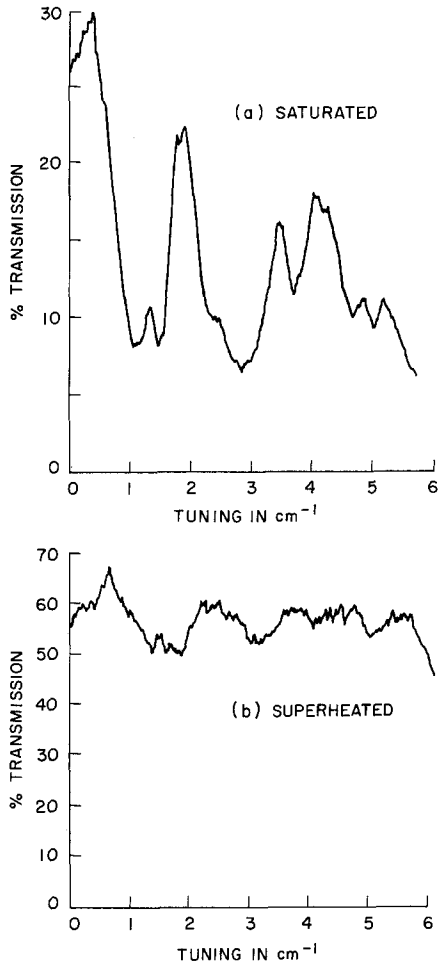

Fig. 4a and b. The transmission near 3500 Å of a laser beam with a linewidth of $0.6\,cm^{-1}$: (a) saturated-vapor mode; (b) superheated-vapor mode. The center of the scan is at 3503.1 Å

of 76 cm, the density of Na_2 molecules is estimated to have been $7\times10^{15}\,cm^{-3}$. The theoretical estimate (Fig. 1) is $1.5\times10^{16}\,cm^{-3}$. This discrepancy is probably due to some slight superheating of the column center due to the split-wick geometry of the cell.

Figure 3b shows the comparable data obtained for the superheated mode of operation. In this case, transmission in the region of the $C-X$ band increased from the 5–10% shown in Fig. 3a to 50–60%. Similarly, transmission on the $D-X$ band increased from approximately 40% to 75%. The lack of improvement in the region of the $B-X$ band seems likely to have been due to larger molecular absorption cross sections for that band. The loss of transmission in the region 3800–4500 Å may be due to scattering since little change occurred between the two modes of operation in the 25% loss measured in that spectral region. The absorption at 3303 and 2853 Å correspond, respectively, to the Na[$3s-4p$] and Na[$3s-5p$] atomic transitions.

At 1300 K, which was the estimated temperature at the center of the inner heater, the theoretical molecular density (Fig. 1) would have been $3\times10^{14}\,cm^{-3}$. This represents a reduction by a factor of 50 of the density of dimers compared to a conventional heat pipe at 20 Torr of Na pressure. However, using the absorption measured for the $C-X$ band, and the measured column length of 59 cm for the superheated mode of operation, an average density of Na_2 molecules of $2.5\times10^{15}\,cm^{-3}$ can be derived. This experimental value includes the contribution from the transition zones of the cell, where the dimer density can be as large as $1.5\times10^{16}\,cm^{-3}$. The measured molecule density can be explained by assuming the theoretically predicted value of $3\times10^{14}\,cm^{-3}$ at the center of the cell and transition zones of 5 cm length, containing saturated vapor.

One of the principal motivations for the development of this cell was to allow population measurements to be made of excited state species produced by x-ray photoionization and photoelectron excitation. Such measurements can be made by monitoring the absorption from an excited state transition. However, this requires a probe beam to pass through the cell without excessive attenuation. In addition, the absorption due to molecules must not exhibit significant frequency dependence over the range of frequencies to be probed.

To test the feasibility of such a measurement in the region of the $C-X$ band, a dye laser, with a linewidth of approximately $0.6\,cm^{-1}$, was passed through the cell and scanned over a $6\,cm^{-1}$ region around 3503 Å. Figure 4 shows the results of this test. In Fig. 4a, for the case, where the cell was operating in the conventional heat-pipe mode, absorptions as deep as 90% are evident with considerable structure over the $6\,cm^{-1}$ region. In contrast, for the superheated cell, the results of Fig. 4b show absorptions of only 40% and negligible structure. It should be noted that, at the operating temperature of the superheated cell (1300 K), Doppler widths in the region around 3500 Å are approximately $0.1\,cm^{-1}$. Consequently, individual rotational structure would not have been resolved by the probe laser.

3. Summary

In summary, the operation of a superheated Na cell, which is suitable for x-ray photoionization experiments, has been described. A significant improvement has been obtained in the transmission in the region of the $C-X$ and $D-X$ bands of Na_2. In addition to the ten-fold improvement of transmission on the $C-X$ band, a negligible frequency dependence

was observed in the residual absorption at 3503 Å, at a resolution of 0.6 cm^{-1}. The cell design outlined here should be equally applicable to operation with the heavier alkali metals K, Rb, and Cs. The simplicity and ease of operation of the design should make it suitable for other applications, such as Raman scattering or harmonic generation, where high densities of relatively molecule-free alkali vapors are of importance [8, 10].

Acknowledgements. The authors would like to express their gratitude for the encouragement of S. E. Harris and to acknowledge helpful discussions with M. Berg, R. W. Falcone, A. L. Harris, and C. B. Harris. This work was supported by the Office of Naval Research and the U.S. Department of Energy.

References

1. J.C. Wang, R.G. Caro, S.E. Harris: Phys. Rev. Lett. **51**, 767 (1983)
2. R.G. Caro, J.C. Wang, R.W. Falcone, J.F. Young, S.E. Harris: Appl. Phys. Lett. **42**, 9 (1983)
3. R.G. Caro, J.C. Wang, J.F. Young, S.E. Harris: Phys. Rev. A (to be published)
4. S.A. Mani, H.A. Hyman, J.D. Daugherty: J. Appl. Phys. **47**, 3099 (1976)
5. S.E. Harris: Opt. Lett. **5**, 1 (1980)
6. J.E. Rothenberg, S.E. Harris: IEEE J. QE-**17**, 418 (1981)
7. D.R. Stull, G.C. Sinke: *Thermodynamic Properties of the Elements*, Adv. Chem. Ser. No. 18 (American Chemical Society, Washington, DC 1956) pp. 188–189
8. R. Wyatt, D. Cotter: Appl. Phys. **21**, 199 (1980)
9. C.R. Vidal, M.M. Hessel: J. Appl. Phys. **43**, 2776 (1972)
10. A.L. Harris, J.K. Brown, M. Berg, C.B. Harris: Opt. Lett. **9**, 47 (1984)
11. R.W.B. Pearse, A.G. Gaydon: *The Identification of Molecular Spectra*, 4th edn. (Chapman and Hall, London 1976) pp. 247–248
12. R.D. Hudson: J. Chem. Phys. **43**, 1790 (1965)