Disk-shaped heat-pipe oven used for lithium excited-state lifetime measurements

Robert W. Boyd, Jack G. Dodd,* Jerzy Krasinski,† and C. R. Stroud, Jr.

Institute of Optics, University of Rochester, Rochester, New York 14627

A metal-vapor spectroscopic oven based on the heat-pipe principle has been constructed in which the heated stainless steel wicks are mounted on disks spaced closely compared with their radii and in which the window is a glass spacer ring separating the disks and permitting optical access to the active volume. The device has been successfully operated using both sodium and lithium as the working fluid. The lithium $4s$ $^2S_{1/2}$ and $5s$ $^2S_{1/2}$ radiative lifetimes were measured with this device, yielding values of 56 ± 1.7 and 99 ± 9 nsec, respectively.

Introduction

An elegant and effective method of generating a well-defined homogeneous metal vapor of uniform and known density, pressure, and temperature has been developed by Vidal and Cooper[1] from an evaporative conductive device known as a heat pipe, described first by Grover et al.[2] Besides making possible the containment of the vapor in pressure equilibrium with an inert gas whose pressure is easily measured externally, the heat-pipe oven isolates the often corrosive vapor from windows, which can be maintained at ambient temperature.

The numerous advantages of the device as an oven for spectroscopic studies have to some extent been offset by the linear geometry that makes Raman and other fluorescence experiments inconvenient, both because weak fluorescence in the forward direction can be difficult to separate from ever-present primary-beam scattering and because some fluorescence properties (e.g., polarization) are inherently functions of angle. The first of these difficulties was overcome by Hessel et al.,[3] who constructed a heat pipe in the shape of a cross. The design presented in this Letter solves the second problem by permitting unrestricted angular access to the active volume without sacrificing any of the advantages of the heat-pipe oven. Also presented in this Letter are the results of a measurement of the fluorescent lifetimes of the $4s$ $^2S_{1/2}$ and $5s$ $^2S_{1/2}$ (denoted by $4s$ and $5s$, respectively, in the following discussion) levels of atomic lithium performed using this device.

Disk-Shaped Heat-Pipe Oven

By reversing the aspect ratio of the conventional heat-pipe oven and exchanging windows and walls, we developed the design shown in Fig. 1. Electrical heating elements (Chromalox CIR 2015) are inserted into copper or silver blocks held in good thermal contact by hard solder to the thin central surfaces of the stainless steel oven. Recesses machined into the insides of the top and bottom plates make it possible to hold circular 5-ply stainless-steel mesh wicks flush with the surrounding surfaces by short flat-headed stainless steel screws. The wall of the heat pipe is a glass ring about 18 cm in diameter having a 0.635-cm-diameter polished window to allow entrance of a collimated (dye-laser) beam. The observing angle is unrestricted, and it is quite practical to design experiments in which this angle is scanned.

During operation, the vapor occupies a disk-shaped region at the center of the oven whose pressure is determined by the external gas (argon) pressure and whose diameter is determined by this pressure and by the input power to the heaters. The boundary between vapor and buffer gas is well defined, as in the conventional heat pipe. Heat is transferred by the vapor, which condenses at the edges of the active region and flows back as a liquid under capillary action toward the warmer center. The heat is removed by annular cooling coils through which water circulates. Unlike the conventional heat-pipe oven whose heat-transfer rate is independent of the length of the active region, our design has an effective heat-transfer rate proportional to the radius of the active region, which may be as much as several centimeters.

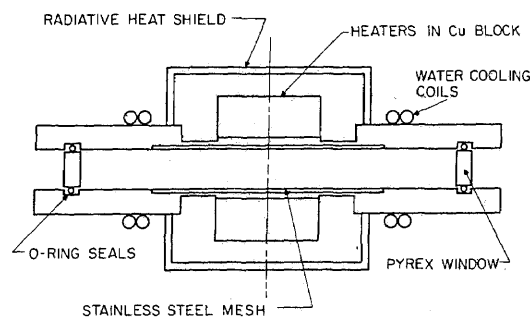The design shown in Fig. 1, with 1 kW of electrical

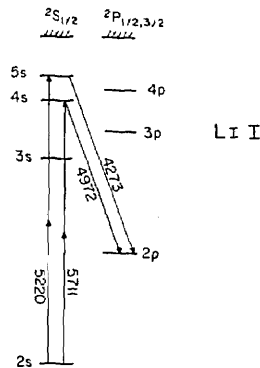Fig. 1. Cross-sectional view of the disk-shaped heat-pipe oven.

Fig. 2.   Energy-level diagram for LiI, showing excitation and fluorescent wavelengths.

heating, has proved reliable up to temperatures of 550°C, at which the vapor pressure of Na is approximately 10 Torr. In order that the 900°C temperature needed to obtain 10 Torr of lithium vapor could be reached, the design had to be modified by removing the electrical heaters and heating the stainless steel plates with propane–air torches. Temperatures as high as 1200°C were obtained by using this method of heating.

Because the upper and lower surfaces of the oven can be electrically isolated from each other, it is possible to apply an electric field to the vapor. In fact, the intent to study the Stark effect in alkali vapors led us to this design.

Lithium Lifetime Measurement

Measurements of the lifetimes of the excited states of an atomic system provide a sensitive test of atomic theory since predicted lifetimes depend strongly on calculated wave functions. A recent theoretical treatment is that of Lingard and Nielson,[4] in which excited-state lifetimes for lithium are calculated by using the Coulomb approximation[5] and in which extensive references to earlier theoretical work can be found.

A number of methods have been used for the measurement of lithium lifetimes. Andersen et al.,[6] Bickel et al.,[7] and Buchet et al.[8] have used beam-foil excitation in the measurements of numerous lithium lifetimes. In particular, Bickel et al. measure the 4s lifetime as 48 ± 2 nsec, and Buchet et al. measure the 4s and 5s lifetimes as 55.8 ± 2.8 and 113 ± 6 nsec, respectively. Karstensen et al.[9] have measured lithium lifetimes by using the delayed-coincidence method following electron-impact excitation, whereas Schulze-Hagenest et al.[10] have measured the 3d lifetime by using laser excitation of a fast beam of excited lithium atoms. These methods permit cascading atoms to populate the upper fluorescing level and thus increase its lifetime. Corrections must be applied to cancel this effect, and these corrections are a possible source of systematic error. The 2p lifetime was determined by Brog et al.[11] from the radiative linewidth of a single hyperfine component in a lithium-atomic-beam, level-crossing experiment. Furthermore, Heldt et al.[12] have measured the lifetimes of the 3d, 4d, and 2p levels by time-resolved detection following one-photon or stepwise two-photon excitation

of an atomic beam. In summary, the 4s and 5s lifetimes have been measured previously[7,8] by using beam-foil techniques, but the work described here is the first reported laser-spectroscopic measurement of these lifetimes.

Experimental Method

In the present study, the lifetimes of the lithium 4s and 5s levels (shown in Fig. 2) were measured by directly exciting one of these levels by a two-photon transition and monitoring the decay of the fluorescence from this level to the 2p level. A uniform vapor of atomic lithium is provided by the disk-shaped heat-pipe oven shown in Fig. 3. The two-photon transition to the 4s level was excited by a 30-kW, f/12, 5711-Å beam from a nitrogen-laser-pumped rhodamine 6G dye laser, whereas the 5s level was excited at 5220 Å by using coumarin 4 as the laser dye. The $ns \rightarrow 2p$ fluorescence was isolated by a narrow-bandpass filter and was detected by a cooled RCA 7265 photomultiplier whose output was sampled by a PAR 162 boxcar averager and displayed on an x–y recorder. Exponential decay times were determined from the slope of a straight-line fit to the data on a semilog plot, as shown for a typical case in Fig. 4.
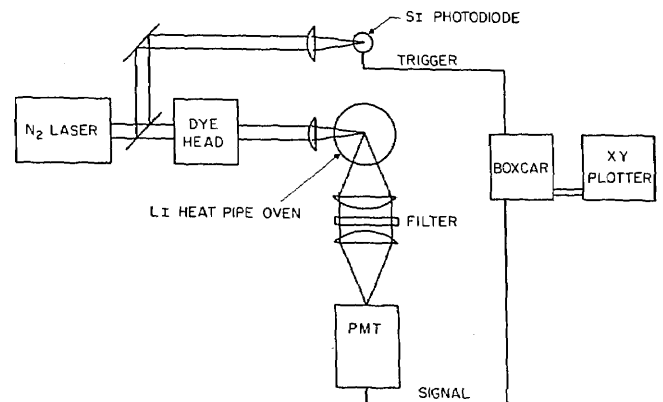

Fig. 3.   Experimental setup used for the lithium lifetime measurements.
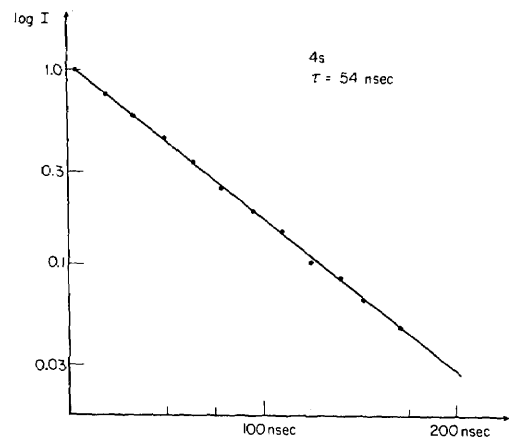

Fig. 4.   Exponential decay of the fluorescence from the 4s level at a lithium partial pressure of 0.11 Torr.
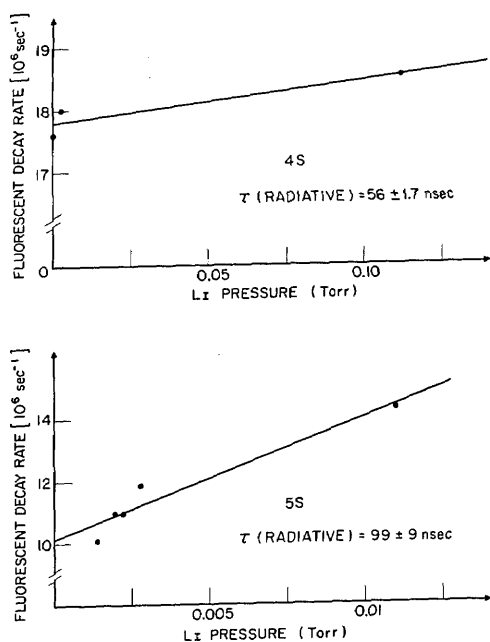
Fig. 5. Fluorescent decay rate versus lithium partial pressure for the 4s and 5s levels. The straight line is a least-squares fit to the data points, and the inferred radiative lifetime is the reciprocal of the vertical-axis intercept of this line.

For lithium pressures of $\gtrsim 1$ Torr, our cell was operated in the true heat-pipe mode, and for these pressures the two-photon-induced fluorescence was clearly visible by eye. To avoid spurius results caused by collisional effects, our lifetime measurements were made at much lower pressures with the heat pipe operated as a conventional cell with an argon buffer gas whose pressure was held sufficiently low ($\simeq 0.1$ Torr) that the measured lifetimes showed no buffer-gas pressure dependence. The temperature of the vapor was measured by a nichrome–tungsten thermocouple placed in the working volume directly above the lithium-wetted wick, and the lithium partial pressure was inferred from this temperature by assuming that the vapor was saturated. Decay rates were measured for several lithium partial pressures, and, as shown in Fig. 5, the true radiative decay rate was determined by extrapolating the measured rates to zero pressure. The radiative lifetimes measured by this procedure are in good agreement with the theoretical predictions of Lingard and Nielsen.[4] In particular, the measured 4s lifetime is 56 ± 1.7 nsec, whereas the theoretical value is 56.6 nsec; the measured 5s lifetime is 99 ± 9 nsec, whereas the theoretical value is 103 nsec.

Conclusions

A heat-pipe oven of novel geometry permitting efficient collection of fluorescent radiation has been used in the measurement of the radiative lifetimes of the lithium 4s and 5s levels. Resonant two-photon excitation of the fluorescing level eliminates the possibility of errors resulting from cascades. The measured lifetimes show a small dependence on lithium partial pressure, which probably is due to collisional de-excitation. By extrapolation to zero pressure, true radiative lifetimes are obtained, and these values are in good agreement with theoretical predictions. This method is applicable to other levels of lithium and, because of the high temperature achievable in the oven (1200°C), to many other atomic systems. Initial results show that strong fluorescent decay signals can be obtained for lithium S and D levels up to $n = 10$. Measured lifetimes are likely to show interesting pressure dependences for such high levels.

* The permanent address of J. G. Dodd is Physics Department, Colgate University, Hamilton, New York 13346.
† The permanent address of J. Krasinski is Institute of Experimental Physics, Warsaw University, Hoza 69, 00-681 Warsaw, Poland.

References

1. C. R. Vidal and J. Cooper, J. Appl. Phys. 40, 3370 (1969).
2. G. M. Grover, T. P. Cotter, and G. F. Erickson, J. Appl. Phys. 35, 1990 (1964).
3. M. M. Hessel and P. Jankowski, J. Appl. Phys. 43, 209 (1972).
4. A. Lindgard and S. E. Nielsen, At. Data Nucl. Data Tables 19, 533 (1977).
5. D. R. Bates and A. Damgaard, Philos. Trans. R. Soc. London Ser. A 242, 101 (1949).
6. T. Anderson, K. A. Jessen, and G. Sorensen, Phys. Lett. 29A, 384 (1969).
7. W. S. Bickel, I. Martinson, L. Lundin, R. Buchta, J. Bromander, and I. Bergstrom, J. Opt. Soc. Am. 59, 830 (1969).
8. J. P. Buchet, A. Denis, J. Desesquelles, and M. Dufay, C. R. Acad Sci. Paris, 265 B, 471 (1967).
9. F. Karstensen and J. Schramm, J. Opt. Soc. Am. 57, 654 (1967).
10. D. Schulze-Hagenest, H. Harde, W. Brand, and W. Demtroder, Z. Phys. A282, 149 (1977).
11. K. C. Brog, T. G. Eck, and H. Wieder, Phys. Rev. 153, 91 (1967).
12. J. Heldt and G. Leuchs, Z. Phys. A291, 11 (1979).