Measurement of lithium and sodium metastable quartet atoms in a hollow-cathode discharge

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We report the measurement, in a pulsed hollow-cathode discharge, of metastable quartet atoms of Li and Na. By using a tunable probe laser, population densities of 3×10^{10} atoms cm^{-3} and 10^{11} atoms cm^{-3} were measured in the Li$(1s2s2p)^4P^\circ$ and Na$(2p^53s3p)^4D_{7/2}$ levels, respectively. These levels are candidates for energy storage for extreme-ultraviolet lasers.

Recent proposals[1–3] for extreme-ultraviolet (XUV) lasers are based on energy storage in core-excited levels of alkali atoms that are metastable[4] against both autoionization and radiation. A tunable, picosecond-time-scale laser would transfer the stored population to a radiatively allowed level in the doublet series.[5,6] Figure 1 is an energy-level diagram for such a system in neutral Li. The storage level Li$(1s2s2p)^4P^\circ_{5/2}$ lies 57.4 eV above the Li$(1s^22s)^2S$ ground level. Since this is the lowest quartet level of the atom, there is no lower configuration into which the atom may decay in the absence of a spin–spin interaction.

In this Letter we report the measurement, in a pulsed hollow-cathode discharge, of metastable populations in the $1s2s2p\ ^4P^\circ$ level of Li at 57.4 eV and in the analogous $2p^53s3p\ ^4D_{7/2}$ level of Na at 33.1 eV. The measurements were performed in a modified version of a pulsed hollow-cathode discharge described earlier.[7] Metastable populations were determined by measuring the absorption of laser probe beams on strong quartet–quartet transitions at 371.4 nm ($1s2s2p\ ^4P^\circ$–$1s2p^2\ ^4P$) in Li, and at 388.3 nm ($2p^53s3p\ ^4D_{7/2}$–$2p^53s3d\ ^4F^\circ_{9/2}$) in Na,[8] as indicated in Figs. 2(a) and 2(b). Fine-structure splitting is small in Li ($\sim$1 cm^{-1}) and was not resolved in these absorption measurements. The population we report in Li therefore includes contributions from the $J = 1/2, 3/2, 5/2$ states.

Figure 3(a) shows the design of the hollow-cathode discharge. A 5-cm-diameter stainless-steel tube contains the metal vapor and serves as the discharge anode. Inside and concentric with this outer tube is a 30-cm-long stainless-steel tube of 1.9-cm diameter, which functions as the cathode. The cathode is suspended by three 0.95-cm-diameter support rods, which enter through sidearms spaced 10.16 cm apart and attached to the main anode body. The structure is heated to 900°C for Li and 530°C for Na and is operated in a heat-pipe mode at a vapor pressure of approximately 6 Torr. This corresponds to ground-level densities of 5×10^{16} and 8×10^{16} cm^{-3} of Li and Na, respectively.

A high-voltage pulse (typically 5 kV) was applied to the cathode through the conducting support rods, producing a plasma inside the cathode [Fig. 3(b)]. Four evenly spaced 0.9-cm-diameter holes in the cathode ensured a uniform plasma along the cathode length. The discharge was pulsed at 10 Hz by using a thyratron switch to connect a 200-nF capacitor to the hollow cathode through a low-inductance electrode geometry. Typical voltage and current waveforms are shown in Ref. 9. Maximum instantaneous power into the discharge occurred approximately 200 nsec after breakdown, at which time the cathode-to-anode voltage was 2000 V and the current was 1500 A. An advantage of this side-fed hollow-cathode discharge configuration is its ability to scale to arbitrary cathode lengths by employing more sidearm structures.

The laser probe beam at 371.4 nm for measuring the Li$(1s2s2p)^4P^\circ$ population was obtained by frequency doubling the output of a pulsed dye laser and had a linewidth of approximately 1.2 cm^{-1}. So adequate sensitivity could be obtained, the probe beam was

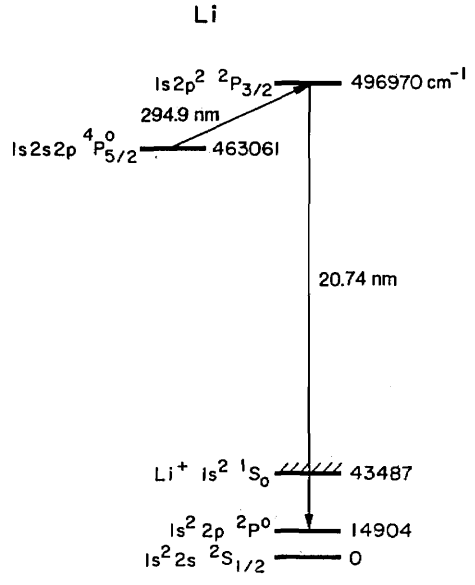

Fig. 1. Energy-level diagram for a 20.7-nm laser in neutral Li.

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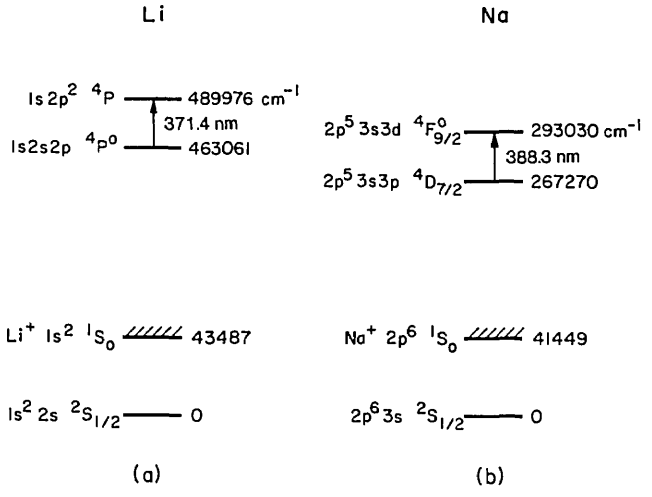

Fig. 2. Energy-level diagrams for quartet population measurement in Li and Na.

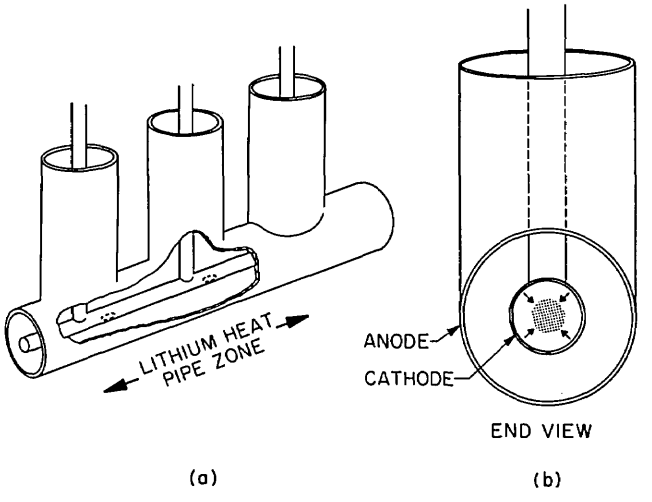

Fig. 3. Diagram of the hollow-cathode discharge. The end view shows the discharge plasma filling the inside of the cathode, with arrows representing high-energy electrons emitted from the cathode.

multipassed through the discharge by using an optical cavity formed by two Al mirrors of 4-m radius of curvature spaced 180 cm apart. Eight passes of the probe through the discharge produced an effective path length in the plasma of 240 cm.

Population densities were determined by using the curve-of-growth method. Experimental absorption curves were fitted to computer-generated curves resulting from the convolution of the probe-laser line shape with the atomic Voigt profile. The generated curves included the fine and hyperfine splittings of the upper and lower transition levels,[10,11] a Doppler width of 0.15 cm^{-1}, and a laser probe width of 1.2 cm^{-1}. At the Li quartet population densities observed, the absorption measurement is relatively insensitive to the (collision-broadened) Lorentzian width of the transition. The total oscillator strength for the Li $1s2s2p$ $^4P^o$–$1s2p^2$ ^4P transition[11] is $f = 0.36$. Figure 4 shows the experimental (solid curve) and computer-generated absorption curves for Li. The best fit implies a Li$(1s2s2p)^4P^o$ population of $(3 \pm 1) \times 10^{10}$ atoms/cm^3. The error bound comes from intensity fluctuations of the laser and uncertainty in the hyperfine splitting.

The population of the metastable Na$(2p^53s3p)^4D_{7/2}$ level was measured in a similar manner. In this case, the fine-structure components are distinct, and hyperfine splitting is negligible. The laser probe width at 388.3 nm was 0.5 cm^{-1}, and the Doppler width was 0.1 cm^{-1}. An absorption oscillator strength of $f = 0.37$ for the $2p^53s3p$ $^4D_{7/2}$–$2p^53s3d$ $^4F_{9/2}$ transition was inferred from the spontaneous decay time of 7.6 nsec measured by Berry *et al.*[8] The calculated Lorentzian width that resulted from Stark and collision broadening was 0.008 cm^{-1}, which is probably correct to within an order of magnitude. Allowing for this uncertainty in the actual Lorentzian linewidth, the best fit to the experimental absorption line shape was obtained for a Na$(2p^53s3p)^4D_{7/2}$ population of $N = (6 \pm 4) \times 10^{10}$ atoms/cm^3. Subsequently, we have intensified the hollow-cathode discharge by decreasing the cathode diameter from 1.9 to 0.95 cm. This has resulted in an increased Na$(2p^53s3p)^4D_{7/2}$ population of $N = (1.0 \pm 0.7) \times 10^{11}$ atoms/cm^3. We also observe absorption at 386.6 nm, which we believe corresponds to the transition $2p^53s3p$ $^4D_{7/2}$–$2p^53s3d$ $^4F_{7/2}$.

To obtain some normalization on the magnitude of these populations, we note that the pulsed hollow cathode, when operating under similar conditions, produces 3×10^{11} ions/cm^3 in the metastable Li$^+(1s2s)^1S$ level.[7] Population in this level is produced by electron ionization of $1s^2$ electrons with a cross section of about 2×10^{-18} cm^2. This cross section is probably about an order of magnitude larger than the spin-exchange cross section, which is responsible for quartet production.[4] Since steady-state metastable population is determined by the ratio of excitation rate to deexcitation rate, we view the results reported in this

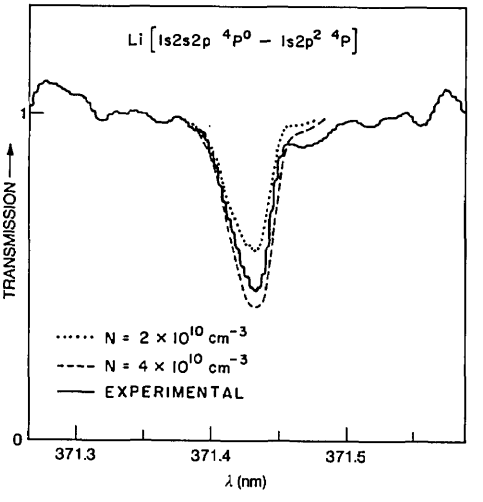

Fig. 4. Experimental and computer-generated absorption curves for Li$(1s2s2p)^4P^o$ population density.

Letter as reasonable evidence that the Li$(1s2s2p)^4P°$ and Na$(2p^53s3p)^4D_{7/2}$ levels are not unusually susceptible to electron or ion deexciting collisions. A recent direct measurement of the Li$(1s2s2p)^4P°$ metastability in the presence of about 10^{15} electrons and ions per cubic centimeter yields a lifetime of about 3 nsec.[12] This is within a factor of 2 of the lifetime of the metastable $Li^+(1s2s)^1S$ level, as measured under similar conditions.[13]

If we assume that the lower level of the XUV transition is empty, and that 50% of the populations measured here are transferred to the upper level of the radiating XUV transition (Fig. 1), then the gain at 20.7 nm in Li and at 37.5 nm in Na would be about 3 and 25% per meter of discharge length. More-intense discharges or reflectors, which allow multipassing, will be necessary to demonstrate an XUV laser.

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