Extreme-ultraviolet fluorescence from core-excited levels of neutral rubidium

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Strong extreme-ultraviolet fluorescence originating from core-excited autoionizing levels of neutral Rb is observed. Radiative branching ratios approaching unity are inferred from the radiative yields. Long autoionizing lifetimes and fast radiative rates make these levels promising candidates for extreme-ultraviolet laser systems.

In general, core-excited levels that lie above a continuum can decay by autoionization. Although there are exceptions in light elements for which a contradiction between orbital angular momentum and parity conservation may prohibit autoionization, in heavier elements the spin–orbit interaction leads to the breakdown of these selection rules. For these elements, excitation of electrons from the outermost closed shell results in levels that generally autoionize at least 100 times faster than they radiate. Because of their small radiative branching ratios, extreme-ultraviolet (XUV) fluorescence is seldom observed from such levels.

We recently reported the development of a tunable-laser-based technique that permits the measurement of long autoionizing times.[1,2] In the course of studying neutral Rb we measured lifetimes of 310 and >500 psec, respectively, for the $4p^55s6s\ ^2P_{1/2}$ and $^4P_{3/2}$ levels. Using the RCN/RCG atomic physics code,[3] we calculated radiative lifetimes of 8.3×10^{-10} and 2.9×10^{-9} sec for the XUV transitions at 75.7 and 76.2 nm ($4p^55s6s \rightarrow 4p^66s$), respectively, which originate from these levels. It was thus expected that we should be able to observe fluorescence on these transitions. The measurement of this fluorescence, together with an estimate of radiative branching ratios, is reported here.

The laser-induced fluorescence technique that we use is based on the properties of the Rb $4p^55s5p\ ^4S_{3/2}$ core-excited level. This level, termed quasi-metastable,[4,5] serves both as a reservoir of population and as an energy reference level.[6] Because of its 2.3% admixture with $4p^55s5p\ ^2P$, it radiates on the 82.4-nm $4p^55s5p\ ^4S_{3/2} \rightarrow 4p^65p\ ^2P_{3/2}$ transition and also permits laser coupling to odd-parity core-excited doublet and quartet levels.

The quasi-metastable level is excited by charge transfer from $Rb^+\ 4p^55s\ ^3P_1$ ions,[7] which are in turn produced by laser-generated x rays. As is shown in Fig. 1, a tunable dye laser is used to transfer quasi-metastable atoms to other, potentially radiating, levels in the core-excited manifold. As the laser is tuned through a transition, the quasi-metastable population is transferred to the radiating target level, resulting in a depletion of 82.4-nm fluorescence and the appearance of laser-induced fluorescence at a wavelength λ from the target level. The amount of laser-induced fluorescence is the intensity of the λ radiation with the laser tuned on line center, $I_\lambda^{(\mathrm{on})}$, minus the intensity with the laser tuned far off line center, $I_\lambda^{(\mathrm{off})}$. Similarly, the amount of laser-depleted fluorescence is the intensity at 82.4 nm with the laser tuned on line center, $I_{82.4}^{(\mathrm{on})}$, minus the intensity with the laser tuned off line center, $I_{82.4}^{(\mathrm{off})}$. The measured quantity in this experiment is the ratio of the laser-induced fluorescence to laser-depleted fluorescence:

$$R_i = \frac{I_\lambda^{(\mathrm{on})} - I_\lambda^{(\mathrm{off})}}{I_{82.4}^{(\mathrm{off})} - I_{82.4}^{(\mathrm{on})}}. \tag{1}$$

R_i is the relative fluorescent yield of the ith level and is used to infer the radiative branching ratio of the level, as described below. The uncertainty in the measurement of R_i is ±25%, and it is mainly due to the background noise of the signals.

The experimental apparatus is shown in Fig. 2. Soft x rays are produced by 150-mJ, 7-nsec pulses of 1.06-μm radiation focused through the Rb vapor onto

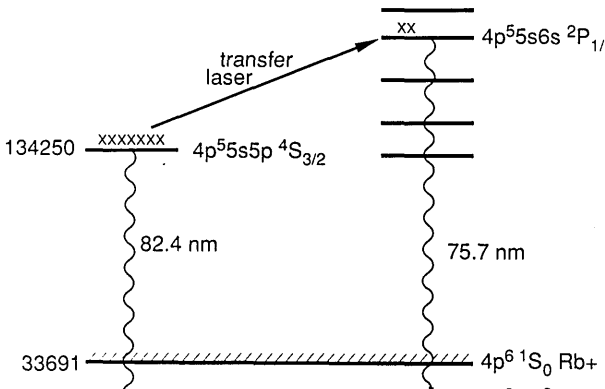

Fig. 1. Partial energy-level diagram of Rb showing the laser-induced fluorescence technique.

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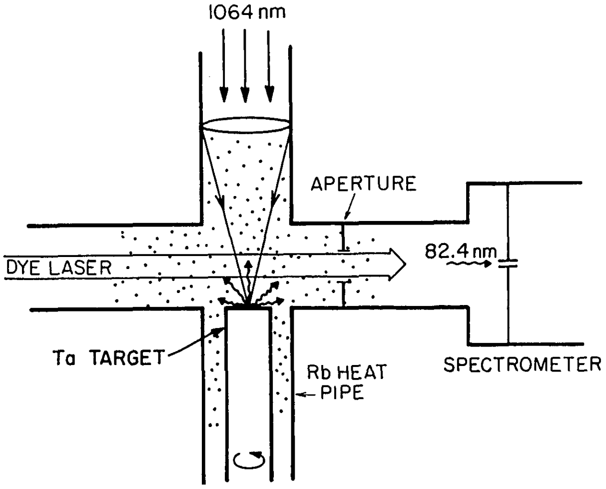

Fig. 2. Schematic of the experimental apparatus.

a Ta target. The dye laser has a pulse width of 5 nsec and is incident upon the Rb vapor 20 nsec after the beginning of the 1.06-μm pulse. The detector time gate used to sample the XUV fluorescence has a width of 15 nsec and is opened at the time of arrival of the dye-laser pulse. It is important that the width of this time gate be small compared with the measured decay time (>40 nsec) of the quasi-metastable level.

Figure 3 shows the results of the laser-induced fluorescence technique applied to the core-excited transition at 553.4 nm. This wavelength is identified in Ref. 1 as belonging to the $4p^5 5s5p\ {}^4S_{3/2} \rightarrow 4p^5 5s6s\ {}^2P_{1/2}$ transition. The upper level, $4p^5 5s6s\ {}^2P_{1/2}$, lies 152 315 cm^{-1} above the ground level. Figure 3(a) shows the 82.4-nm fluorescence intensity as a function of dye-laser wavelength as the laser is tuned through this transition. At line center of the transition, an 80% depletion of the 82.4-nm fluorescence from the $4p^5 5s5p\ {}^4S_{3/2}$ quasi-metastable level is observed. The laser energy is well above the saturation energy of the transition; failure of the depleted signal level to reach the baseline is due mainly to imperfect overlap of the depleted and viewed volumes. In Fig. 3(b) the spectrometer is tuned to 75.7 nm and the signal level recorded as a function of dye-laser wavelength. At line center of the transfer transition, laser-induced fluorescence is observed. The wavelength of the induced fluorescence, 75.7 nm, corresponds to 132 100 cm^{-1} and identifies the lower level as $4p^6 6s\ {}^2S_{1/2}$. The XUV transition at 75.7 nm is therefore identified as $4p^5 5s6s\ {}^2P_{1/2} \rightarrow 4p^6 6s\ {}^2S_{1/2}$. The relative fluorescence yield of the level is $R_i = 0.54$.

The relative fluorescent yield of the ith level can be used to infer its radiative branching ratio, $(\mathrm{BR})_i$. If fluorescence is collected during a time gate of length T, where T is long compared with the lifetime of level i and short compared with the lifetime of the quasi-metastable level, then

$$(\mathrm{BR})_i \equiv \frac{A_i}{\Gamma_i} = A_{\mathrm{qm}} T R_i, \tag{2}$$

where Γ_i is the total decay rate of level i and A_{qm} is the radiative decay rate of the quasi-metastable level. This relation is obtained by integration of the rate equations for the populations in the two levels and also assumes that the time gate opens after the excitation of the quasi-metastable level has ceased.[7]

Since the radiative rate of the quasi-metastable lev-

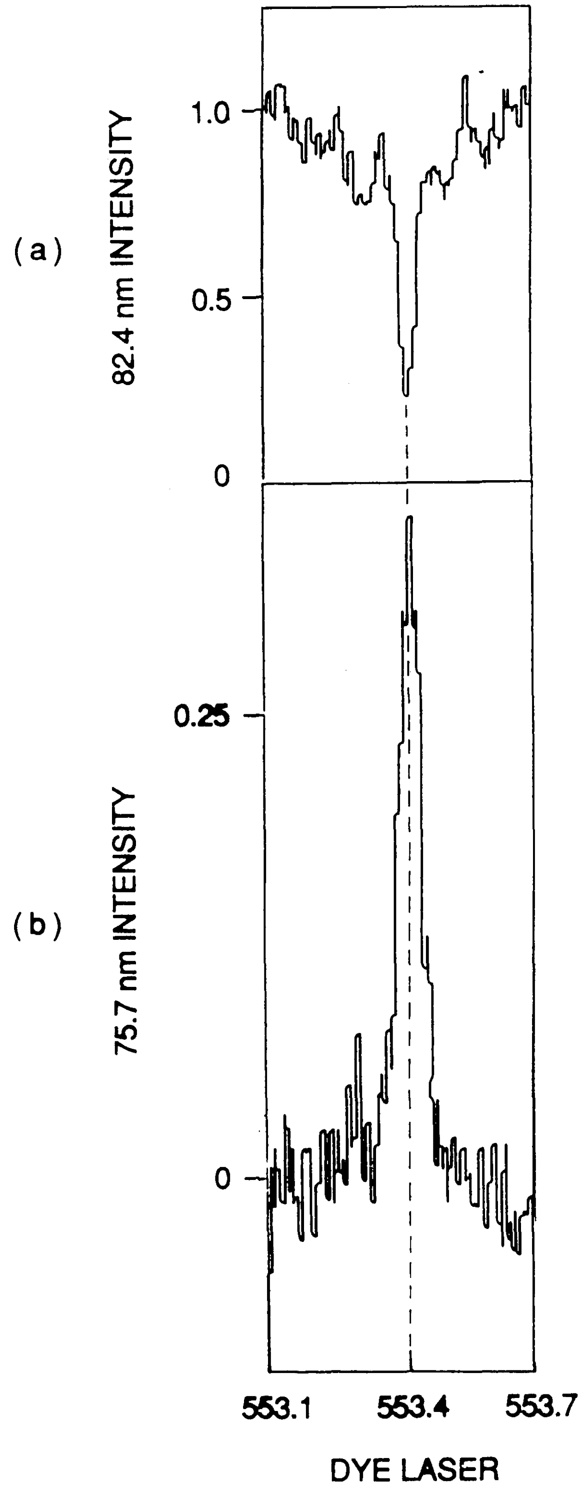

Fig. 3. Experimental scans showing (a) the depletion of 82.4-nm fluorescence by the dye-laser transfer at 553.4 nm and (b) laser-induced fluorescence at 75.7 nm.

Table 1. Summary of Experimental and Calculated Results

	XUV Transition Wavelength			
	75.7 nm	76.2 nm	77.2 nm	82.4 nm
Upper Level	$4p^55s6s\ ^2P_{1/2}$	$4p^55s6s\ ^4P_{3/2}$	$4p^54d(^3D)5s\ ^2D_{3/2}$	$4p^55s5p\ ^4S_{3/2}$
Lower Level	$4p^66s\ ^2S_{1/2}$	$4p^66s\ ^2S_{1/2}$	$4p^64d\ ^2D_{5/2}$	$4p^65p\ ^2P_{3/2}$
Γ_i (sec^{-1}) (measured)[a]	3.2×10^9	3.8×10^8	5.0×10^{10}	–
R_i (measured)[b]	0.54	1.90	0.04	–
Radiative branching ratio	0.21	0.74	0.02	–
Γ_i (sec^{-1}) (calculated)	2.2×10^9	5.3×10^{10}	5.9×10^{10}	–
A_i (sec^{-1}) (calculated)	1.2×10^9	3.4×10^8	4.1×10^8	2.6×10^7
A_i/Γ_i (calculated)	0.55	0.006	0.007	–
σ_g (cm^2)	3.6×10^{-13}	1.1×10^{-13}	1.3×10^{-13}	9.6×10^{-15}

[a] Ref. 2.

[b] Uncertainty is ±25%.

el, A_{qm}, has not, to date, been measured, we proceed by using a calculated value. The calculation is performed by the RCN/RCG code,[3] which computes the positions and transition strengths of the levels of the following configurations: $4p^65s$, $4p^66s$, $4p^64d$, $4p^55s5p$; $4p^65p$, $4p^55s^2$, $4p^55s6s$, $4p^54d5s$, $4p^54d^2$, $4p^55p^2$. A scale factor of 0.77 is used for all radial integrals. This calculation gives $A_{\mathrm{qm}} = 2.6 \times 10^7$ sec^{-1}. For a gate width of $T = 15$ nsec we obtain $\mathrm{BR}_i = 0.39R_i$. For the 75.7-nm transition this gives a radiative branching ratio of 0.21.

XUV fluorescence was found to originate from three of the fifteen levels that were identified previously.[1,2] Table 1 summarizes the measurements and also compares branching ratios obtained from the measured R_i values with branching ratios calculated by using the RCN/RCG code. In all cases the transition wavelengths are consistent with the identifications and lifetimes of Refs. 1 and 2.

The large branching ratio of the $4p^54d(^3D)5s\ ^2D_{3/2}$ level can be attributed to the *LS* selection rule prohibiting autoionization for a level of even orbital angular momentum and odd parity. The long autoionizing times for the $4p^55s6s$ levels must be attributed to a cancellation in the radial integral. We note that Silfvast *et al.*[8] have noted a similar cancellation for autoionization of the Cd^+ $4d^95s6s\ ^2D_{5/2}$ level and measured a decay time of 450 psec.

Because these transitions terminate on excited levels of the valence manifold and because the oscillator strengths between the quasi-metastable level and their upper levels are quite large, these transitions provide near-prototype systems for store-and-transfer lasers.[9] The calculated gain cross sections of these transitions are included as the last row of Table 1.

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