Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium

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This Letter describes a new technique for obtaining level positions, linewidths, autoionizing times, and oscillator strengths of core-excited levels and transitions. The technique uses a tunable laser to deplete the population of a radiating core-excited level, as other levels within the core-excited manifold are accessed. Level positions and linewidths are ascertained to within 1.0 cm^{-1} accuracy, and autoionizing times whose Lorentzian linewidths lie beneath the combined Doppler-hyperfine structure are measured.

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Knowledge of the locations and autoionizing times of core-excited autoionizing levels is important for the understanding of many physical processes, as well as for its applicability to the generation of coherent extreme ultraviolet and soft x-ray radiation. Traditionally, the positions of these levels have been measured by ejected-electron spectroscopy, and by absorption spectroscopy either from the ground level or from laser-prepared valence levels.[1] More recently, Cooke *et al.*[2] and Bloomfield *et al.*[3] have used multistep and multiphoton excitation to prepare doubly excited column-II atoms, and to measure their linewidths and autoionizing times.[4] Also, Holmgren *et al.*[5] have used a tunable laser to transfer population from a metastable level in neutral Na to target levels, and by observing the resulting fluorescence, have defined much of the quartet manifold.

In this Letter we describe experimental results on a new technique[6,7] which allows the measurement of autoionizing times, linewidths, relative level positions, and transition oscillator strengths of core-excited levels. The technique is based on the large radiative rates, relative to their autoionizing rates, of certain levels that recently have been termed quasimetastable.[8] Each of the column-I metals and the column-II alkali-metal-like ions has one or two such levels which radiate strongly in the extreme ultraviolet (xuv) to levels in the valence structure.[9] These quasimetastable levels lie at, or near, the bottom of the core-excited manifold of both parities. In this technique we use laser-produced x rays to excite impulsively a quasimetastable level, and following this excitation monitor the generated xuv radiation. A tunable transfer laser is then scanned over the region where autoionizing levels are expected to be present. When a level is encountered, the quasimetastable population is transferred to it, depleting the xuv fluorescence. The shape and position of the depleted signal, as functions of transfer-laser wavelength, determine the position and linewidth of the autoionizing level. By using a saturation technique which is described below, we measure the autoionizing times of levels which are sufficiently long lived that their Lorentzian autoionizing linewidth lies beneath their combined Doppler-hyperfine profile.

Figure 1 shows a partial energy-level diagram of neu-

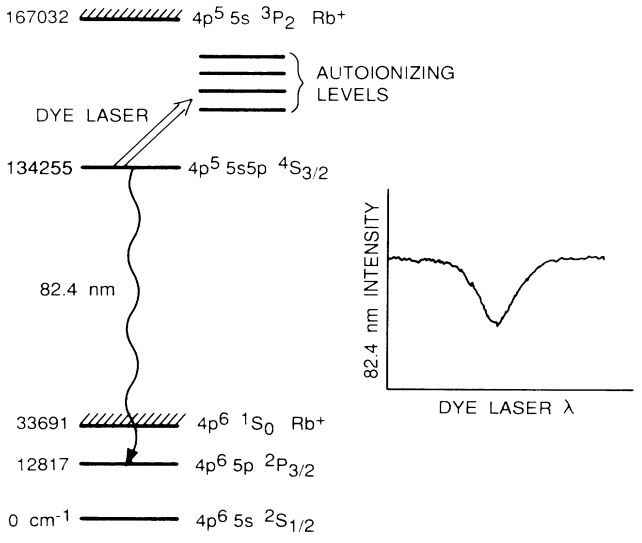

FIG. 1. Partial energy-level diagram of Rb showing the depletion spectroscopy technique.

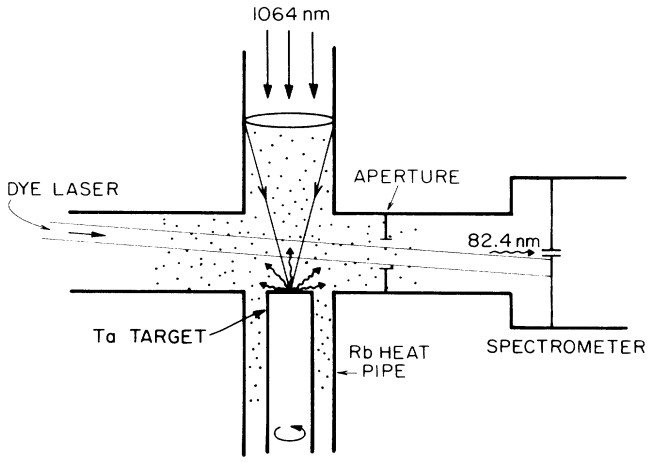

FIG. 2. Experimental schematic.

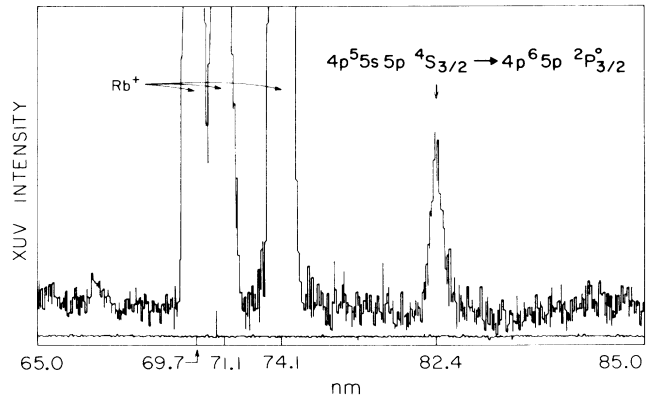

FIG. 3. xuv emission spectrum of Rb.

tral rubidium. In Rb, the lowest quasimetastable levels[9] are the even-parity $4p^5 5s 5p\ ^4S_{3/2}$ level which radiates at 82.4 nm, and the odd-parity $4p^5 4d 5s\ ^4P_{5/2}$ level which radiates at 85.2 nm. In this work we monitor the 82.4-nm radiation and therefore access odd-parity levels with $J = \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$. Figure 2 shows a schematic of the experimental arrangement. A 7-ns, 100-mJ pulse of 1064-nm Nd-doped yttrium-aluminum-garnet radiation is focused onto a tantalum target to a 100-μm-diameter spot and therefore to a power density of 2×10^{11} W cm^{-2}. The plasma created by this laser radiates soft x rays into the surrounding region, which is filled with Rb vapor at a density of 2×10^{16} cm^{-3}. The x rays ionize the vapor, producing $Rb^+(4p^5 5s)$ and hot electrons. Both of these species excite the $Rb(4p^5 5s 5p\ ^4S_{3/2})$ quasimetastable level, the first by charge transfer, and the second by direct electron excitation. Figure 3 shows an emission scan of soft-x-ray excited Rb, taken 20 ns after the end of the 1064-nm laser pulse. Over a 10-nm-wide spectral region, the 82.4-nm quasimetastable radiation is the dominant spectral feature. This allows use of 1-mm-wide spectrometer slits and therefore relatively good collection efficiency.

The transfer beam is a tunable dye laser, pumped by the same 1064-nm laser that provides the x-ray excitation. Typically the dye laser has an energy of 10 mJ cm^{-2} and a linewidth of 0.25 cm^{-1}, and is scanned at a rate of 1 cm^{-1} per second.

Figure 4 shows the 82.4-nm intensity as a function of the frequency of the tunable laser, in the vicinity of the quite wide $4p^5 4d(^1P)5s\ ^2P_{3/2}$ level. We place this level at 19794 cm^{-1} above the quasimetastable level, and measure a linewidth of 34 cm^{-1}. This width corresponds to an autoionizing time of 1.6×10^{-13} sec. The identification of this, as well as other levels, is achieved by comparison of the measured lifetime, transition oscillator strength, and level position with the predictions of the RCN/RCG atomic physics code,[10] and also by comparison with the xuv absorption data of Mansfield[11] and the ejected-electron data of Pejcev *et al.*[12]

Figure 5(a) shows the line shape of the narrow $4p^5 5s 6s\ ^4P_{5/2}$ level which we place at 16339 cm^{-1} rela-

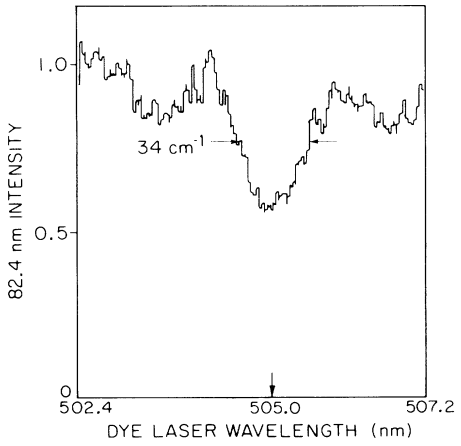

FIG. 4. Depletion scan of the transition $4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 4d(^1P)5s\ ^2P_{3/2}$. Laser energy is equal to 1.0×10^{-3} J cm^{-2}.

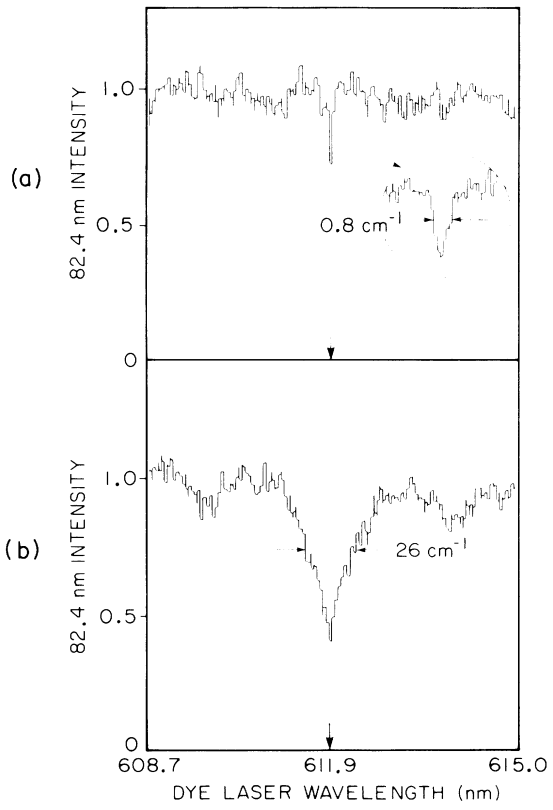

FIG. 5. Depletion scan of the transition $4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 5s 6s\ ^4P_{5/2}$ showing Lorentzian wing power broadening. (a) Laser energy is equal to 6.8×10^{-6} J cm^{-2}. (b) Laser energy is equal to 1.2×10^{-2} J cm^{-2}.

TABLE I. Relative level energies, linewidths, autoionizing times, and designations.

Relative[a] energy (cm^{-1})	Linewidth (cm^{-1})	Autoionizing time (ps)	Tentative designation
14654	0.7	> 20	$4p^5 4d(^3D)5s\ ^2D_{3/2}$
16339	0.8	53	$4p^5 5s 6s\ ^4P_{5/2}$
17060	0.7	> 530	$4p^5 5s 6s\ ^4P_{3/2}$
18064	0.8	> 440	$4p^5 5s(^1P)6s\ ^2P_{1/2}$
19161	0.8	33	$4p^5 5p^2\ ^4D_{5/2}$
19794	34	0.16	$4p^5 4d(^1P)5s\ ^2P_{3/2}$

[a]Level energies are relative to the $4p^5 5s 5p\ ^4S_{3/2}$ level. On the basis of Mansfield's (Ref. 11) position of $4p^5 4d(^3D)5s\ ^2D_{3/2}$, we place the $4p^5 5s 5p\ ^4S_{3/2}$ level at 134255 ± 5 cm^{-1}. A recent measurement by Reader (Ref. 13) places it at 134250.1 ± 0.4 cm^{-1}.

tive to the quasimetastable level. Since in this case the 0.8-cm^{-1} linewidth is on scale with the combined Doppler–hyperfine profile, its reciprocal is not equal to the autoionizing time. In this case we use a saturation technique which allows the measurement of Lorentzian linewidths which lie beneath the combined Doppler–hyperfine-transfer laser linewidth. The technique is closely related to a method demonstrated by Cooke, Bhatti, and Cromer,[4] and makes use of the fact that the transition probability in the far wings of a power-broadened Voigt profile varies as the oscillator strength and Lorentzian linewidth of the transition, and is independent of the Doppler–hyperfine-transfer laser linewidth. Figure 5(b) shows 82.4-nm intensity as a function of laser detuning for the same level ($4p^5 5s 6s\ ^4P_{5/2}$) as that of Fig. 5(a), but this time at an incident laser energy density that is about 2000 times larger. Using computer-generated power-broadened line shapes, we compare calculated values with the experimental results for a number of laser detunings and intensities. For this level we measure a Lorentzian linewidth of 0.10 cm^{-1}, which corresponds to an autoionizing time of 53 ps.

We have also measured the oscillator strength of each of the transitions from the $4p^5 5s 5p\ ^4S_{3/2}$ quasimetastable level to the accessed odd-parity autoionizing levels. This measurement is made by tuning of the transfer laser to line center and recording of the laser energy density at which the depletion curve reaches $\exp(-1)$ of its maximum depth. This saturation energy density is inversely proportional to the absorption cross section, and therefore, together with the measured (total) linewidth, determines the oscillator strength of the transition.

Table I lists the energies relative to the $4p^5 5s 5p\ ^4S_{3/2}$ quasimetastable level, the measured (low laser intensity) linewidths, and the autoionizing times of each of the accessed levels. Table II lists the oscillator strength of each of the accessed transitions. Oscillator strength measurements are estimated to be accurate to a factor of 2, and in turn introduce an uncertainty of the same order of magnitude into the Lorentz width measurements. The uncertainty in relative level positions is about 1 cm^{-1}. The absolute energy of the accessed levels depends on that of the initial quasimetastable level. Use of Mansfield's[11] position for the $4p^5 4d(^3D)5s\ ^2D_{3/2}$ level places the $4p^5 5s 5p\ ^4S_{3/2}$ at 134255 ± 5 cm^{-1}. This is within the error bounds of the value given by Mendelsohn *et al.*[9] A recent measurement by Reader[13] places the $4p^5 5s 5p\ ^4S_{3/2}$ level at 134250.1 ± 0.4 cm^{-1}. The > signs on the autoionizing times in Table I are the result of either our inability to measure very narrow Lorentz linewidths, or the possibility that the widths have a collisional or radiative component.

In summary, we have demonstrated a new method for measuring relative positions of core-excited levels, core-excited linewidths, and autoionizing times to unprecedented accuracy. The method can be used to access core-excited levels of both parities and with J values that do not allow measurement from the ground level. The large signals which are obtained by the x-ray excitation

TABLE II. Transition wavelengths and oscillator-strength measurements. The tabulated quantity gf is the absorption oscillator strength f multiplied by the degeneracy of the quasimetastable level, $g = 4$.

Transition	Wavelength (nm)	Oscillator strength (gf) Experiment	Calculation
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 4d(^3D)5s\ ^2D_{3/2}$	682.24	0.003	0.002
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 5s 6s\ ^4P_{5/2}$	611.91	1.0	1.131
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 5s 6s\ ^4P_{3/2}$	585.99	0.2	0.195
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 5s(^1P)6s\ ^2P_{1/2}$	553.44	0.03	0.029
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 5p^2\ ^4D_{5/2}$	521.76	0.07	0.057
$4p^5 5s 5p\ ^4S_{3/2} \rightarrow 4p^5 4d(^1P)5s\ ^2P_{3/2}$	505.02	0.1	0.431

method allow scan rates of at least 1 cm^{-1} per second, thereby making this an extraordinarily versatile and powerful technique. Further experiments may allow the observation of Rydberg series, interferences between autoionizing levels, continuum repulsion, and dressed-level effects characteristic of laser-induced autoionization.[14]

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[1]T. J. McIlrath and T. B. Lucatorto, Phys. Rev. Lett. **38**, 1390 (1977).

[2]W. E. Cooke, T. F. Gallagher, S. A. Edelstein, and R. M. Hill, Phys. Rev. Lett. **40**, 178 (1978).

[3]L. A. Bloomfield, R. R. Freeman, W. E. Cooke, and J. Bokor, Phys. Rev. Lett. **53**, 2234 (1984).

[4]W. E. Cooke, S. A. Bhatti, and C. L. Cromer, Opt. Lett. **7**, 69 (1982).

[5]D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris, Phys. Rev. A **31**, 677 (1985).

[6]S. E. Harris, J. F. Young, A. J. Mendelsohn, D. E. Holmgren, K. D. Pedrotti, and D. P. Dimiduk, in *Laser Spectroscopy VII*, edited by T. W. Haensch and Y. R. Shen, Springer Series in Optical Science Vol. 49 (Springer-Verlag, New York, 1985), pp. 162–165.

[7]K. D. Pedrotti, Ph.D. dissertation, Stanford University, 1985 (unpublished), and "Extinction Spectroscopy: A Novel Laser Spectroscopic Technique" (to be published).

[8]S. E. Harris, D. J. Walker, R. G. Caro, and A. J. Mendelsohn, Opt. Lett. **9**, 168 (1984).

[9]A. J. Mendelsohn, C. P. Barty, M. H. Sher, J. F. Young, and S. E. Harris, Phys. Rev. A **35**, 2095 (1987).

[10]Robert D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California, Berkeley, 1981), Sects. 8-1, 16-1, and 18-7.

[11]M. W. D. Mansfield, Astrophys. J. **183**, 691 (1973).

[12]V. Pejcev, D. Rassi, K. J. Ross, and T. W. Ottley, J. Phys. B **10**, 1653 (1977).

[13]Joseph Reader, "Accurate Energies for the $4p^5 5s 5p\ ^4S_{3/2}$ and $4p^5 5s 4d\ ^4P_{5/2}$ Levels of Neutral Rubidium (Rb)" (to be published).

[14]K. Rzazewski and J. H. Eberly, Phys. Rev. Lett. **47**, 408 (1981).