Oscillator Strength Measurements of Even-Parity Autoionizing Resonances by Combined Synchrotron-Radiation–Laser Excitation

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We have obtained oscillator strengths for transitions between a laser-excited initial state and autoionizing final states. In the case of sodium, a laser was used to populate the $3p$ initial state, and synchrotron radiation was used to excite the autoionizing resonances. These transitions are of the type $2p^6 3p\ ^2P \rightarrow 2p^5(^2P)3s3p(^{1,3}P)^{2,4}S,^{2,4}P,^{2,4}D$; they decay to $2p^6\ ^1S + e$. The sum of the oscillator strengths for all the observed transitions between the $2p^6 3p$ initial-state and the $2p^5 3s3p$ final-state configurations was found to be equal to 0.22(4).

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We have measured, for the first time, oscillator strengths (OS) for transitions involving even-parity autoionizing levels by a hybrid synchrotron-radiation–laser experiment. These measurements are the result of the development of a new technique to study ionization processes in atoms that are in excited states. As an illustrative example, we present measurements in sodium of the oscillator strengths for transitions between a laser-excited initial $2p^6 3p$ configuration and autoionizing levels of a $2p^5 3s3p$ final-state configuration. Measurements of the OS for inner-shell transitions to autoionizing levels in sodium are of special interest[1] because the quartet terms of the core-excited levels are metastable and are possible upper states for an extreme-uv laser.

Autoionization has been observed in almost every molecule, atom, or ion studied.[2] Most of these studies have been made on transitions that are dipole allowed from the ground state. For example, lasers and collisional-excitation processes have been used recently to study[3] the profile of the very narrow autoionizing resonances between the $^2P_{3/2}$ and $^2P_{1/2}$ ionization thresholds in neon. Lasers are now used to produce, via stepwise outer-shell excitations, levels of the same parity as the ground state.[4] In previous experiments to observe inner-shell excitations in laser-excited alkali metals and alkaline earths, a pulsed continuum source has been used with pulsed lasers to measure, in photoabsorption, the energy position of inner-shell autoionizing resonances in elements like barium[5] and sodium.[6]

In this paper we use a new method[7] to extract oscillator strengths from the vacuum ultraviolet (vuv) photoemission spectrum of laser-excited sodium. Laser radiation is combined with synchrotron radiation from the Anneau de Collisions d'Orsay (ACO). The monochromatic output of a toroidal grating monochromator (TGM)[8] is focused by a toroidal output mirror onto a weakly collimated sodium beam emanating from a furnace mounted on the axis of a cylindrical mirror analyzer (CMA). Electrons emitted from the source volume at an angle of 54°44′ (the "magic angle") are energy analyzed. For an isotropic initial state, the signal produced by electrons ejected at this angle is independent of the polarization of the radiation and the angular distribution of the electrons and is proportion-

al to the cross section.[9] This arrangement has been used to study atoms[10] and molecules[11] in the ground state. To extend this research to excited states,[7] we have mounted a ring dye laser so that its beam traverses the CMA in a direction perpendicular to the CMA axis at the focus of the synchrotron radiation.

Typical photoelectron spectra from the CMA are shown in Fig. 1. In the upper panel is shown a spectrum with the laser off and the monochromator set at a photon energy, $h\nu_1$, equal to 32.73 eV. Photons of twice the first-order photon energy (65.46 eV) are also transmitted through the monochromator and are the only photons able to photoionize the $2p$ shell electrons observed as peaks 1 and 3 in the figure. With the laser

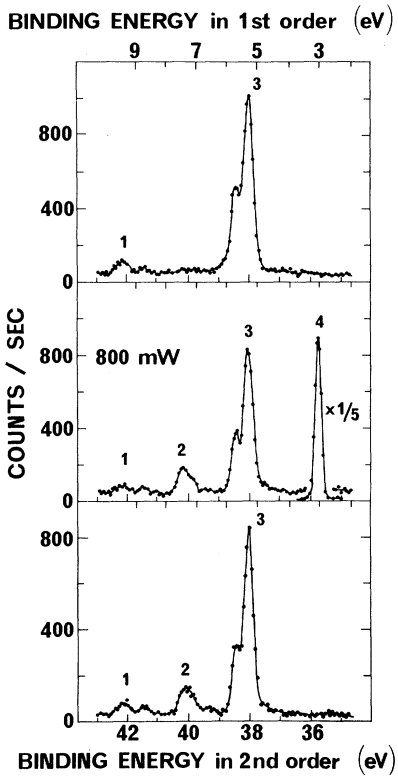

FIG. 1. Top: A photoelectron spectrum of sodium taken at $h\nu_1 = 32.73$ eV with the laser turned off. Peaks 1 and 3 are due to ionization of the $2p$-shell electrons by photons of energy equal to twice $h\nu_1$ or 65.46 eV. The appropriate binding energy (E_B) scale is at the bottom of the figure and is computed from the kinetic energy (E_{kin}) according to $E_B = 2h\nu_1 - E_{\text{kin}}$. Middle: A spectrum taken at $h\nu_1 = 32.73$ eV with the laser turned on. Peak 2 is due to photoionization of the $2p$-subshell electron with the $3s$ electron excited to a $3p$ orbital leaving an ion state $2p^5 3p$. Peak 4 is due to autoionization of the $2p^5(^2P)3s3p(^1P)^2D$ resonance. The appropriate E_B scale for peak 4 is at the top of the figure and is computed from the E_{kin} according to $E_B = h\nu_1 - E_{\text{kin}}$. Bottom: A spectrum taken off resonance at $h\nu_1 = 32.50$ eV with the laser on. Peak 4 disappears, but the peak due to the $2p$ ionization of the laser-excited atom is still present.

switched on (middle panel) photoelectron peaks 2 and 4 appear. Peak 2 is due to the photoionization of $2p$-shell electrons with the $3s$ electron laser-excited to a $3p$ orbital (excitation energy 2.10 eV); peak 4 is due to autoionization from the discrete state resonantly excited by $h\nu_1$, which is the final state of the transition

$$2p^6 3p + h\nu_1 \rightarrow 2p^5(^2P)3s3p(^1P)^2D_{5/2,3/2}.$$

The evidence of this decay is manifested by the presence of the intense peak at the binding energy, E_B, of 3.03 eV (the E_B of a $3p$ electron). When the monochromator is tuned off resonance (lower panel of Fig. 1) at 32.5 eV, peak 4 disappears because the nonresonant $3p$ photoionization cross section is very small, but the peaks 1, 2, and 3 due to photoionization of $2p$-shell electrons by second-order radiation remain. The peak at a E_B of 40 eV, corresponding to the direct photoionization of $2p$-subshell electrons in laser-excited sodium, occurs at a higher E_B than that of ground-state sodium because the $3p$ electron interacts weakly with the $2p$ shell. In an atom like sodium where the outer shell is weakly coupled to the inner-shell electron, one expects, and indeed one calculates,[12] that at the same final-state energy the inner-shell photoionization cross section of an atom with an excited outer electron is the same as the photoionization cross section of the atom in the ground state. We confirmed the validity of these calculations by measuring the changes in the $2p$ photoelectron intensity with the laser on and off.

We now show that the oscillator strength of an autoionizing resonance can be measured from the information contained in Fig. 1. The area[13] $N_{h\nu}$ of the photoelectron peak (2 in Fig. 1) for $2p$ ionization of the laser-excited atoms is proportional to the product of the excited-state density, n_{3p}, and the photoionization cross section, $\sigma(h\nu)$:

$$N_{h\nu} = KI_{h\nu}E_{h\nu}n_{3p}\sigma(h\nu), \tag{1}$$

where K is the spectrometer constant, $I_{h\nu}$ is the photon flux integrated over the spectrometer bandpass ΔE at energy $h\nu$, and $E_{h\nu}$ is the kinetic energy of the photoelectron. Similarly, the integrated area N_R of the electron peak (4 in Fig. 2) for the autoionizing resonance is given by

$$N_R = 110K'\frac{I_R}{\Delta E_R}E_R n_{3p}\int_{\Delta E_R}\frac{df_R}{d\epsilon}d\epsilon, \tag{2}$$

where the symbols have the same meaning as before and are taken at a photon energy corresponding to the resonance energy. The quantity K' is the product of the spectrometer constant and the branching ratio for autoionization. The cross section and the monochromator bandpass are expressed in units of 10^{-18} cm^2 and electronvolts, respectively.[14] In our experiment the monochromator bandpass was much larger than the width of the resonance. Thus the integral over the oscillator strength density $df_R/d\epsilon$ in Eq. (2) can be re-

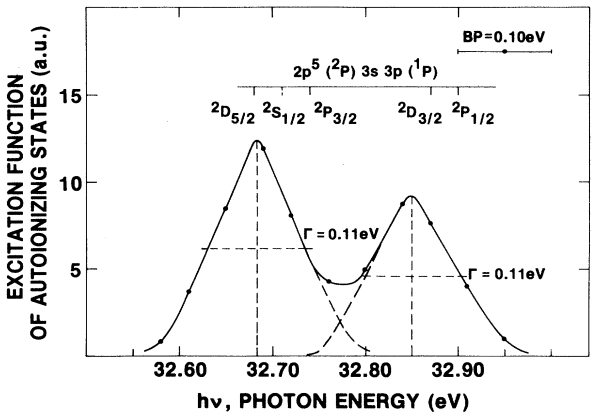

FIG. 2. Excitation function as a function of photon energy for a TGM bandpass (BP in figure) of 0.10 eV. The scale of the ordinate has arbitrary units (a.u.). The configuration and terms of the autoionizing resonance in the spectral range shown in the figure are indicated according to Ref. 7. The energy position of the $2p^5(^2P)3s3p(^1P)^2D_{3/2}$ term differs by 0.02 eV from that of Ref. 7. The width of the structure, $\Gamma = 0.11$ eV, includes the resonance width plus the TGM bandpass.

placed by the oscillator strength of the resonance f_R. Solving Eq. (1) and Eq. (2) for f_R yields

$$f_R = 0.0091 \frac{K}{K'} \frac{I_{h\nu}}{I_R} \frac{E_{h\nu}}{E_R} [\sigma(h\nu)\Delta E_R] \frac{N_R}{N_{h\nu}}. \quad (3)$$

All the quantities in Eq. (3) have been measured in the experiment except the partial $2p$ cross section for ground-state sodium and the autoionization branching ratio, K'/K. The partial cross section was obtained by combination of the results from Refs. 10 and 15, and the autoionization branching ratio was assumed equal to 1. This assumption is valid generally for strict LS coupling, which applies to low-Z elements.

The photon polarization generally induces an alignment in the atom, which has the effect that the initial state for the absorption of the second photon is not randomized. The angular dependence of the photoionization cross section generally includes terms proportional to Legendre polynomials of greater than second order.[16] The presence of higher-order Legendre polynomials in the expression for the cross section destroys the proportionality between the counting rate and the cross section. In our experiment, however, we are working with a beam whose diameter is about 5 mm and whose density is greater than 10^{12} atoms/cm^3. At these densities the alignment induced by the laser is zero because trapping of the laser radiation in the dense vapor destroys the directionality of the radiation,[17] thus rendering the intermediate $3p$ state isotropic.

When the $2p$ electron is excited to the $3s$ orbital, the parent configuration $3s3p$ couples through the exchange interaction to form $^{1,3}P$ terms that are separated by about 1.3 eV. These two terms in turn couple with the $2p^5\,^2P_{3/2,1/2}$ core to form terms of the type $^{2,4}D$, $^{2,4}P$, and $^{2,4}S$ and form two groups of excitation states. The $2p^5(^2P)3s3p(^3P)^2S_{1/2}$ level is isolated from both groups. This resonance and each group associated with the ^3P or ^1P parent was scanned to produce an excitation curve. Figure 2 is the excitation curve that was obtained for the group of resonances that belongs to the $2p^5(^2P)3s3p(^1P)$ configuration.

The widths of the dashed-curve peaks in Fig. 2 were adjusted for a best fit to the data. The solid curve is the sum of the dashed curves, which have the mathematical form of the instrument function and are normalized to the peak counting rates. This fit was made on the assumption that the ^2P terms decay radiatively and the observed intensity is due to just the autoionization of the ^2D terms. The even-parity ^2P terms cannot autoionize because autoionization requires the parities of the initial and final state to be the same,[18] and the only final state available, $2p^6(^1S_0)\epsilon p\,^2P$, has odd parity. The ^2S term can autoionize but is too weak to be observed in these measurements.

A similar excitation curve was made for the terms having $2p^5(^2P)3s3p(^3P)$ coupling. In this case most of the strength was in the $^2D_{5/2,3/2}$ levels with the $^2S_{1/2}$ level next strongest and then followed by the $^4P_{5/2}$ level. In strict LS coupling these ^4P levels would not autoionize, but their presence implies mixing with ^2D levels that can autoionize. The quartet manifold has been studied[19] recently. The measurements indicate that radiative decay competes strongly with autoionization. The value of the oscillator strength for each transition is shown in Table I. The sum of the oscillator strengths for all the measured transitions of the $2p^63p \rightarrow 2p^53s3p$ array is 0.22(4). While calculations for the oscillator strengths of the individual transitions of this array have not been published, several calculations have been made[20,21] of the oscillator strength of

TABLE I. Oscillator strengths for autoionizing resonances in laser-excited sodium $2p^63p\,^2P \rightarrow 2p^53s3p$.

Classification	$h\nu$ [a]	$h\nu$ [b]	f_R [c]
$2p^5(3s3p\,^3P)^2S_{1/2}$	31.78	31.77	0.022(4)
$2p^5(3s3p\,^3P)^2D_{5/2}$	31.40	31.40	0.087(12)
$2p^5(3s3p\,^3P)^2D_{3/2}$	31.34	31.34	
$2p^5(3s3p\,^3P)^4P_{5/2}$	31.19	31.19	<0.01
$2p^5(3s3p\,^1P)^2D_{5/2}$	32.68	32.69	0.063(11)
$2p^5(3s3p\,^1P)^2D_{3/2}$	32.85	32.87	0.045(3)
			$\Sigma = 0.22(4)$

[a]Present measurement of resonance energy.
[b]Resonance energy from Ref. 6.
[c]The number in parenthesis is the estimated probable error of the measurement.

the transition $2p^6 \rightarrow 2p^5 3s$ for the neon isoelectronic sequence. The average value of the oscillator strengths of the transitions belonging to the first few members of the sequence is 0.23. The value we obtain for the oscillator strength of the entire transition array is a lower bound, but the close correspondence between our measurements and the calculations suggest that *LS* coupling dominates and that the excited $3p$ electron acts chiefly as a spectator.

We have successfully applied laser-synchrotron hybrid sources to obtain the oscillator strengths of inner-shell atomic transitions in excited atoms. The availability of new storage rings designed for use with undulators could provide a means to extend measurements of this type to more highly excited atoms or to ions produced by pulsed laser sources.

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