OBSERVATION OF IONIZATION OF LASER EXCITED ATOMS BY SYNCHROTRON RADIATION

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We have begun a program to measure oscillator strengths of autoionizing resonances that result from a transition in the VUV between a laser excited initial state and a final state in which a core electron is promoted. These measurements demonstrate a new technique to combine synchrotron radiation, laser pumping, and photoelectron spectroscopy.

Measurements of the energy positions of autoionizing resonances have been honed to a fine art over the past 50 years.[1] Total cross section measurements and the parameters that describe autoionizing resonances have been determined.[2] Most of these studies have been made from the dipole allowed ground state. Recently autoionizing resonances have been observed from excited initial states[3] and from ion initial states.[4] We have heard several talks[5],[6] at this meeting which described some of this type of research. In the measurements to be described in this paper, laser radiation is combined with synchrotron radiation, as shown schematicaly in Figure 1, to study the photoionization from excited initial states to continuum final states or to autoionizing final states. Continuum radiation from the Aneau de Collisions d'Orsay (ACO), which is installed at the Universite de Paris-Sud,in Orsay France, is monochromatized by a toroidal grating monochromator (TGM)[7] and is focused by a toroidal output mirror on to a weakly collimated sodium beam emanating from a furnace mounted on the axis of a cylinderical mirror analyzer (CMA). This electron spectrometer is used to study the kinetic energy distribution of the ejected photoelectrons produced by the interaction of the photon beam with the focused synchrotron radiation.

This arrangement has been used to study atoms[8] and molecules[9] in the ground state.

To extend this research to excited states[10], we have mounted a ring dye laser (maximum intensity at the sodium D lines 9w/cm[2]) so that its beam traverses the CMA in a direction perpendicular to the CMA axis at the focus of the synchrotron radiation. The laser is stabilized to 20 MHz by locking it to the $3^2S_{1/2}(F=2) \rightarrow 3\,^2P_{3/2}(F=3)$ transition in an auxiliary collimated sodium beam also shown schematically Figure 1. The size of the laser beam is adjusted so that it just fills the source volume, V. Electrons emanating from this volume at the magic angle (54°48') can be detected.

We have chosen to determine the oscillator strength of a group of autoionizing resonances in sodium[11] with the same parity as the ground state.

The area $N(h\nu)$ of the photoelectron peak from 2p ionization of the laser-excited atoms is proporational to the product of the excited state density and the photoionization cross section $\sigma(h\nu)$[12]:

$$N(h\nu) = K \cdot I(h\nu) \cdot E(h\nu) \cdot n(3p) \cdot \int_{\Delta E} \sigma(h\nu') \cdot W(h\nu - h\nu') d(h\nu') \qquad (1)$$

where K is the spectrometer constant, $I(h\nu)$ is the photon flux at photon energy $h\nu$; $E(h\nu)$ is th kinetic energy of the photoelectron; $n(3p)$ is the density of laser-excited atoms; ΔE is the monochromator band pass, and W is the photon monochromator window function. Similarly, the integrated area $N(h\nu_R)$, of the photoelectron peak for the autoionizing resonance is given by:

$$N(h\nu_R) = K_R \cdot I(h\nu_R) \cdot E \cdot (h\nu_R) \cdot n(3p) \cdot f_R' \qquad (2)$$

where $I(h\nu_R)$ is the photon flux at the resonance excitation energy, $E(h\nu_R)$ is the kinetic energy of the decay electron and ΔE_R is the monochromator bandpass. In our experiment, the monochromator bandpass (about 0.1eV in first order) was much larger than the width of the resonance, and the integral over the oscillator strength density in Eq(2) can be replaced by the oscillator strength of the resonance. The oscillator strength can be obtained from the ratio of Eq(2) to Eq(1), yielding:

$$f_R = \frac{N(h\nu_R)}{N(h\nu)} \cdot \frac{K}{K_R} \cdot \frac{I(h\nu)}{I(h\nu_R)} \cdot \frac{E(h\nu)}{E(h\nu_R)} \cdot \Delta E \cdot \sigma(h\nu) \qquad (3)$$

This ratio is independent of the excited state density, one of the more difficult quantities to determine experimentally. All the terms on the right of Eq(3) can be determined experimentally, but one can obtain a

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very good approximate value of f_B by assuming $\sigma(h\nu)$, the excited state continuum cross section, is equal to the photoionization for the ground state.[13]

In Figure 3 we plot Eq(3) as a function of photon energy for some autoionizing resonances in sodium vapor. Photoionization cross sections from an excited ground state in lithium and barium have also been obtained.
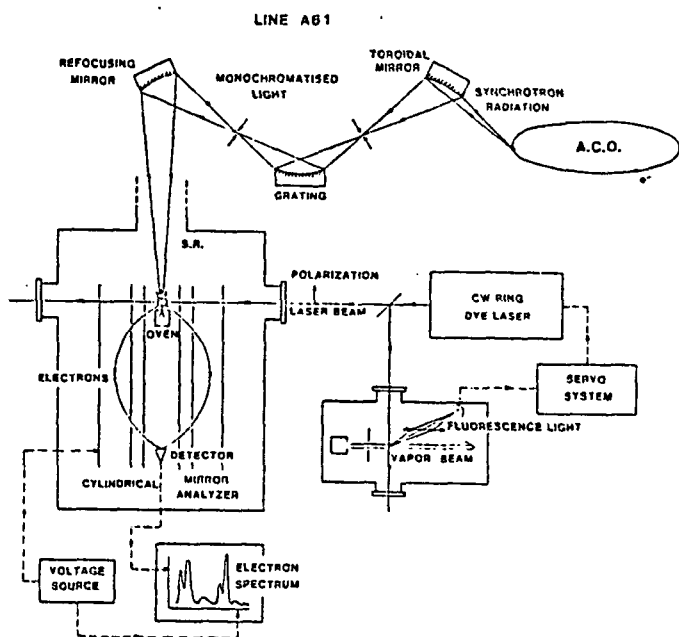

Fig. 1

A schematic representation of the apparatus used for synchrotron radiation studies of laser-eaxcited states. XUV radiation from the ACO storage ring at the Laboratoire pour l'Utilization du Rayonnement Electro-magnêtique (LURE) is monochromatized and is focused on the axis of a cylindrical mirror electron energy analyzer (CMA). An oven located on the CMA axis produces an effusive cloud of sodium which is excited by the stabilized laser beam traversing the CMA perpendicular to its axis. the beam from the ring dye laser (9 Wcm^2 max power) is stabilized to 20 MHz by using fluorescence from the 3 $^2S_{1/2}$(F=2) → $^2P_{3/2}$(F=3) transition in a well collimated sodium beam.
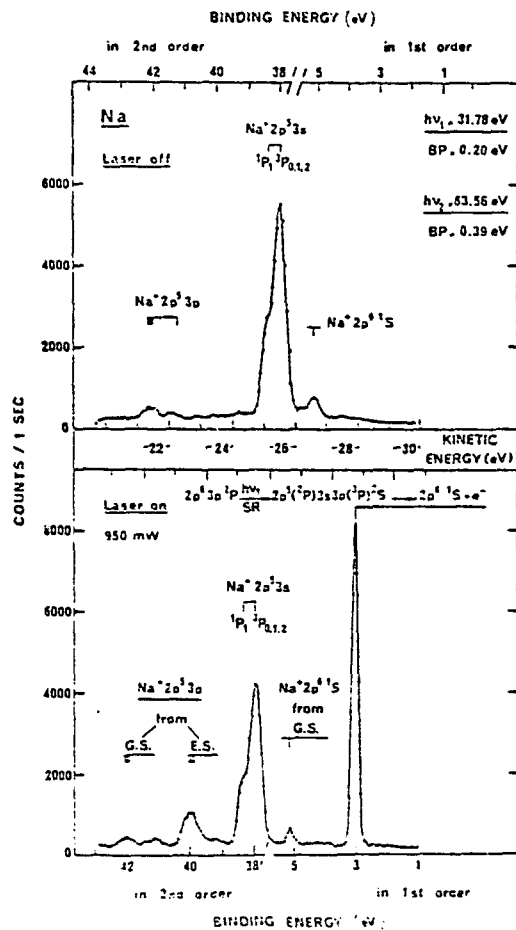

Fig. 2

Photoelectron energy spectrum taken with the TGM set at 31.78 eV. The intense peak at about 29eV kinetic energy (KE) corresponds to the autoionizing decay of the resonance excited by the inner subshell transition indicated in the figure. The small peak at 26.5 eV KE is due to the direct ionization of the 3s electron by the 31.78 eV photons. The structures at 25.5 eV, 22.1 eV, and at 21.4 eV KE are due to the direct ionization of the 2p^{6}3s subshell. by 63.56 eV radiation (2nd order radiation diffracted by the TGM). The structure in the lower frame at 23 eV KE is due to the ionization of a 2p electron in the laser excited sodium

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Fig. 3

Excitation function of laser
excited sodium as a function of
photon energy for a TGM band pass
of 0.10 eV (BP in figure). The
configuration and terms of the
autoionizing resonance in the
spectral range shown in the figure
are indicated according to Ref. 11.
The energy position of the
$2p^5(^2P)3s3p(^1P)^2D_{3/2}$ differs by
0.022 eV from that of Ref. 11. The
width of the structure $\Gamma = 0.11$
includes the resonance widths plus
the TGM bandpass.
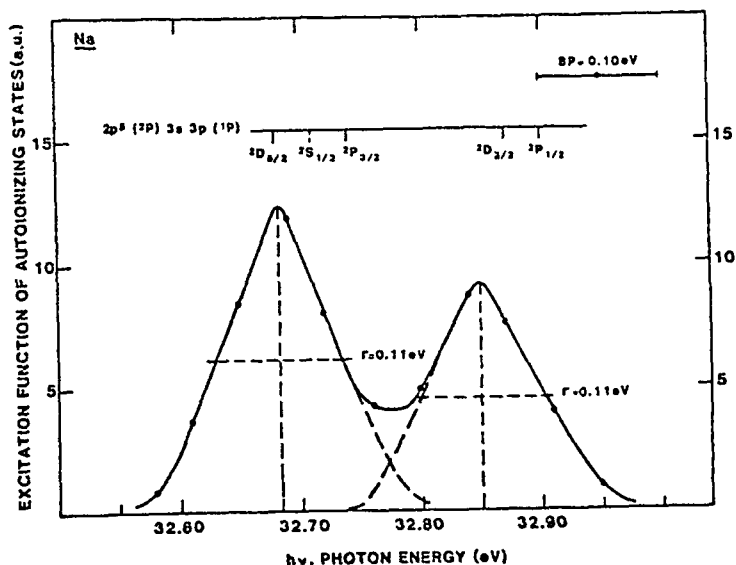
References

1. H. Beutler, Z. Physics 93, 177 (1935); K. Codling, Rep. Prog. Physics 36, 541 (1973).

2. M.O. Krause, F. Cerrina, and T. Fahlman, Phys. Rev. Lett. 50, 118 (1983); J.A.R. Samson, Phys. Lett. 28C 303 (1976).

3. W. Hill, III, K.T. Cheng, W. Johnson, T.B. Lucatorto, T.J. McIlrath, and J. Sugar, Physical Rev. Lett. 49, 1634 (1982).

4. T.J. McIlrath and T.B. Lucatorto, Phys. Rev. Lett. 38, 1390 (1977).

5. M.A. Baig, J.P. Connerade, W.R.S. Garton. J. Hornes, C. Mayhew, G. Noldeke, and K. Sommer, these proceedings.

6. T.J. McIlrath, C. Cromer, V. Kaufman, J. Sugar, W.T. Hill, III, and D. Cooper, these proceedings.

7. P.K. Larsen, W.A.M. van Bers, J.M. Bizau, F. Wuilleumier, V. Schmidt, and D.L. Ederer, Nucl. Instrum. Meth. 195, 245 (1982).

8. S. Krummacher, V. Schmidt, J.M. Bizau, D.L. Ederer, P. Dhez, and F. Wuilleumier, J. Phys. B. 15, 4363 (1982).

9. S. Krummacher, V. Schmidt, F. Wuilleumier, J.M. Bizau, and D.L. Ederer, j. Phys. B. 16, 1733 (1983).

10. J.L. LeGouët, J.L. Picqué, F. Wuilleumier, J.M. Bizau, P. Dhez, P. Koch, and D.L. Ederer. Phys. Rev. Lett. 48, 600 (1982).

11. J. Sugar, T.B. Lucatorto, T.J. McIlrath, A.W. Weiss, Opt. Lett. 4, 109 (1979).

12. M.O. Krause, Chem. Phys. Lett. 10, 65 (1971).

13. T.N. Chang and Y.S. Kim, J. Phys. B. 15, L835 (1982).

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