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Nonlinear Resonant Photoionization in Molecular Iodine

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Strong photoionization spectra have been observed in molecular iodine following laser excitation in the near ultraviolet. The dependence upon laser power, and the structure of the spectra are consistent with three-photon ionization with resonance in an intermediate state excited by two photons.

Ionization of the I_2 molecule has been detected following absorption of three photons of the same frequency at many points in the spectral range 3660 to 3800 Å. The light source was a Molectron DL-300 tunable dye laser pumped by a UV-1000 nitrogen laser, which provided 5-nsec pulses of bandwidth approximately 0.2 Å. The peak power was 14×10^{-5} J/pulse at 3779 Å. The laser beam was focused with a 17-cm–focal-length lens between parallel-plate stainless-steel electrodes in a glass cell containing I_2 vapor at room temperature. The electrodes, separated by 1.1 cm, were maintained at a constant potential difference of 300 V. The frequency of the incident light was continuously variable. The experimental set up is shown in Fig. 1. The voltage across a 1-MΩ resistor in series with the anode was fed into a boxcar integrator and displayed on a chart recorder. The gate width of the boxcar integrator was 50 μsec and the integrating time constant was 0.3 msec, effectively averaging over six laser pulses. Strong bandlike signals

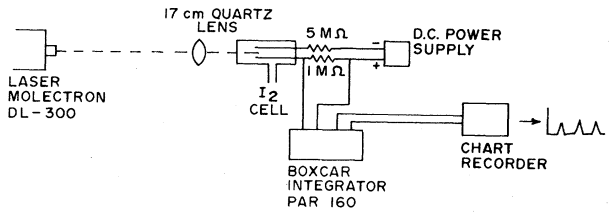

FIG. 1. The apparatus.

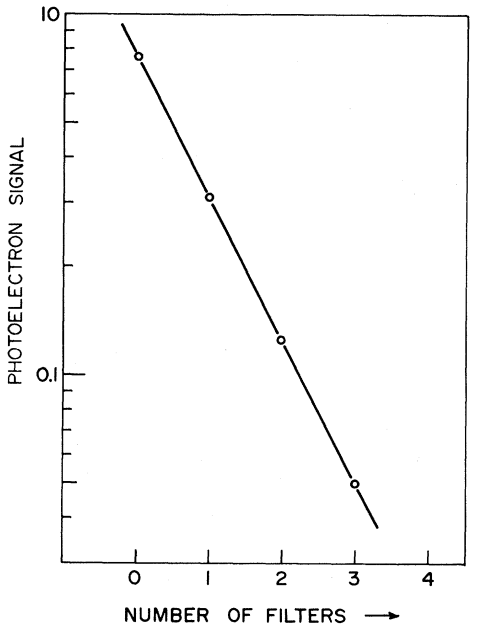

FIG. 2. The output current signal at the laser wavelength 3742 Å as the laser light intensity was varied with calibrated filters of neutral density 0.147.

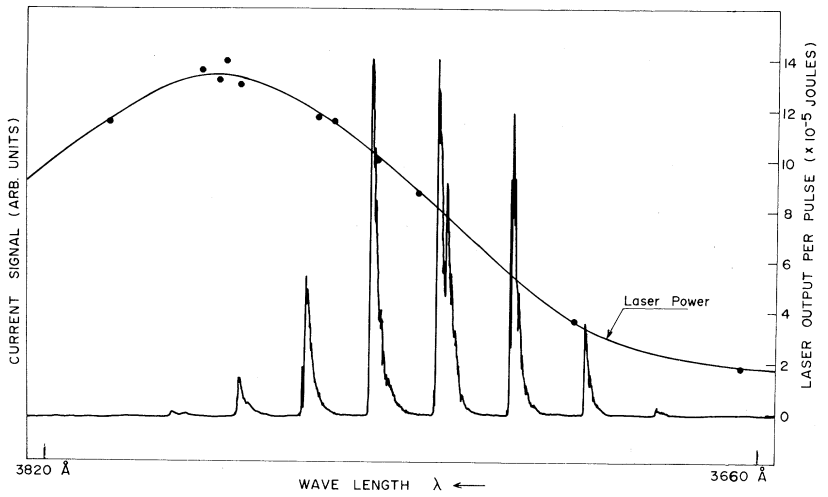

FIG. 3. The observed photoelectron spectrum at relatively fast scan. Full scale signal on the strongest band is about 10^{-8} A. The abscissa represents the laser wavelength in air.

were found as the laser frequency was scanned. The off-resonant voltage was negligible. As single- and double-photon excitation are energetically unable to ionize the I_2 molecules,[1-4] any observed resonance signal must be the result of excitation by three or more photons. The output signal was determined as the light intensity was varied with calibrated neutral-density filters. The data are as shown in Fig. 2. The signal was found to vary as $P^{2.7}$, where P is the laser power. As the electric signal may not be perfectly linearly related to the tripling process because of possible saturation and avalanche breakdown, the variation is consistent with the expected P^3 relation.

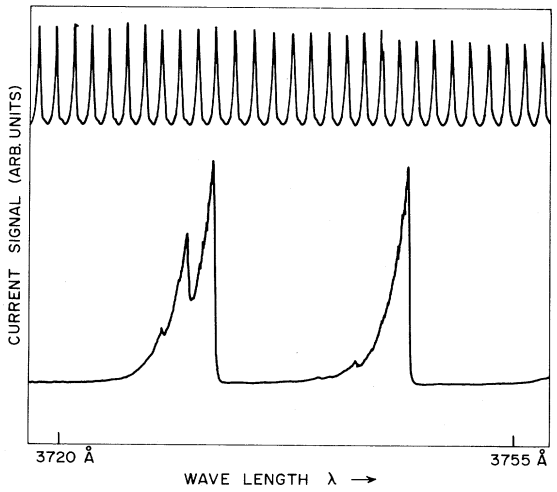

FIG. 4. The lower curve shows two of the photoelectron bands at higher resolution. A small portion of the laser light is fed through a Fabry-Perot interferometer and is detected by a photodiode. The output of the diode is shown above the photoelectron curve to indicate the resolution of the photoelectron bands.

The resulting spectrum, shown in Fig. 3, consists of a series of approximately equally spaced violet-degraded bands. A part of the spectrum is shown at higher resolution in Fig. 4. No resonances can occur for one-photon absorption from the ground state.[1,4] However, two-photon resonance to an intermediate state of symmetry g followed by photoionization would seem to be possible.[5] No bands were found to the red of 3800 Å in spite of the considerably greater laser power in this region. It is reasonable to assign the observed resonances to vibrational bands of two-photon transitions to a I_2 molecular state of g symmetry. Since this intermediate state has not previously been observed (single-photon transition from the ground state to such a state is forbidden), further experimental and theoretical work will be necessary to completely understand and assign the resonances.

In view of the sensitivity and simplicity of the technique described here it is likely that it will have further applications.

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High-Resolution Laser Spectroscopy of the *D* Lines of On-Line Produced ^{21,22,24,25}Na Using a New High-Sensitivity Method of Detection of Optical Resonances

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A polyisotopic sodium beam of $^{21-25}$Na, produced by spallation of Al, was illuminated with a tunable cw dye laser. The atomic beam, analyzed by a six-pole magnet is then ionized and detected by use of a mass spectrometer. The results are as follows: isotopic shifts relative to ^{23}Na, ^{21}Na $=-53.5$ mK, ^{22}Na $=-25.3$ mK, ^{24}Na $=+23.1$ mK, ^{25}Na $=+44.6$ mK; $\mu_I(^{25}\mathrm{Na})=+3.685(22)\mu_N$; b factor in the $P_{3/2}$ state of ^{25}Na $=-16(8)$ MHz.

Half-lives,[1] masses, [2] and other properties[3] of short-lived sodium isotopes have recently been determined by use of a mass spectrometer on-line with an accelerator. As an extension of this work, we wished to determine other static properties of these nuclei. We have thus attempted to measure the hyperfine structure and isotope shift (IS) of the atomic *D* lines of sodium isotopes. The purpose of this paper is to present the first results that have been obtained with a newly developed method that could eventually extend our knowledge on the shape and magnetic properties of nuclei far from stability.

The hfs of the ground state of $^{20-24}$Na has already been measured by different methods[4]; recently a determination of the absolute value of the magnetic moment of ^{25}Na has been published.[5] However the hfs of the excited $3P$ states are unknown for the unstable isotopes and there are no data available for isotope shifts.

This type of measurement on a series of isotopes can give interesting indications on nuclear shapes. The observation of irregularities of the hfs and IS of short-lived Hg isotopes[6] has been especially fruitful in that context. In the case of sodium, however, the same method as in Ref. 6 would fail because of the Doppler broadening, and it is necessary to use one of the new methods of Doppler-free high-resolution spectroscopy.[7-9]

The special requirements of our on-line experiment have in fact led to the development of a variant of one[7] of these methods of detection of optical transitions in an atomic beam.[10] In essence this method rests upon the magnetic detection of the optical pumping which occurs when a laser beam is tuned to the frequency of one of the hyperfine components of the *D* lines. This optical pumping changes the population distribution between the states $m_J=\pm\frac{1}{2}$ of the atoms of an atomic beam, and the change is detected by means of a magnetic filter consisting of a six-pole magnet which focuses the atoms with $m_J=+\frac{1}{2}$ and defocuses the atoms with $m_J=-\frac{1}{2}$.

In the actual experiment, the isotopes of sodium are produced in the spallation of aluminum by 150-MeV protons from the Orsay synchrocyclotron. Since this nuclear reaction results in a mixture of the different isotopes and their optical resonances might overlap, it is necessary to separate them on-line using a mass spectrometer.

A highly efficient target has been developed in order to have the best sodium-isotope production. We have used a molten-metal target in a graphite container. It was loaded with 10 g of high-purity aluminum and heated by alternating current up to 900°C. The yield of the spallation reaction $^{27}\mathrm{Al}(p,3pxn)$Na with 150-MeV protons