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Photoionization from the Selectively Excited $3s3p\ ^1P_1^\circ$ State to the $3p^2\ ^1S_0$ Autoionization Level of Mg I

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The $3s3p\ ^1P_1^\circ$ level of Mg I has been selectively excited by the second harmonic of the output of a high-power, narrow-band, frequency tunable dye laser. The excited-state absorption series $3s3p\ ^1P_1^\circ$-$3snd\ ^1D_2$ has been recorded to $n=24$. A strong autoionization resonance has been obtained at $\lambda=300.9$ nm, with a photoionization cross section of $\sim 5\times10^{-16}$ cm^2, in very good agreement with the values predicted by calculation.

While Bacher showed in 1933[1] that the interaction of $3p^2\ ^1D_2$ with the $3snd\ ^1D_2$ series of Mg I could account for the location of the $3s3d\ ^1D$ term below the corresponding triplet level $3s3d\ ^3D$, the $^1D_2$ and $^1S_0$ displaced terms of the $3p^2$ configuration have not been previously observed. The $^3P_{0,1,2}$ levels of this configuration lie just below the normal ionization limit and give rise to a strong multiplet at $\lambda = 278$ nm[2] in the Mg I spectrum. The presence of $3p^2\ ^1D_2, ^1S_0$ in the continuum will affect the dielectronic recombination rate and the ultraviolet radiation balance in, for example, astrophysical sources. Calculations of the photoionization cross section from the lowest excited singlet level $3s3p\ ^1P_1^\circ$ have been carried out,[3] using the approach of Henry and Lipsky.[4] The continuum functions required for the cross-section matrix elements were obtained from a program of Burke and Moores[5] and the $3s$, $3p$, and $3d$ states of Mg II were included in the representation of the $3p^2\ ^1S_0$ state. The bound $^1P_1^\circ$ function was calculated using a configuration interaction program of Hibbert.[6] Each configuration had the same ($1s^2$, $2s^2$, $2p^6$) core, and the valence-electron functions for the ten configurations were the Slater orbitals

$$(3s, 3p),\ (3p, 3d),\ (4s, 3p),\ (4p, 3d),\ (4f, 3d),$$
$$(3s, 4p),\ (3p, 4d),\ (4s, 4p),\ (4p, 4d),\ (4f, 4d),$$

optimized with respect to the energy. For these bound and continuum functions the photoionization cross section for the $3p^2\ ^1S_0$ resonance was found to have maximum values, at $\lambda = 297.7$ nm, of 12.9×10^{-16} cm^2 in the dipole-length approximation and 12.5×10^{-16} cm^2 in the dipole-velocity approximation.

Higher series members and autoionization levels are normally studied in absorption, but transitions from the Mg I $3s^2\ ^1S_0$ ground state to $^1S_0$, $^1D_2$ states are forbidden. Fortunately, the new possibilities for atomic[7-10] and molecular[11] spectroscopy opened up by the development of high-power, frequency-tubable dye lasers include the selective excitation of atomic singlet states to population densities sufficient to obtain, in absorption, transitions to levels not reached from the ground state. Thus, stimulated electronic Raman scattering from excited alkali vapors[12] has been obtained. Extended excited-state absorp-

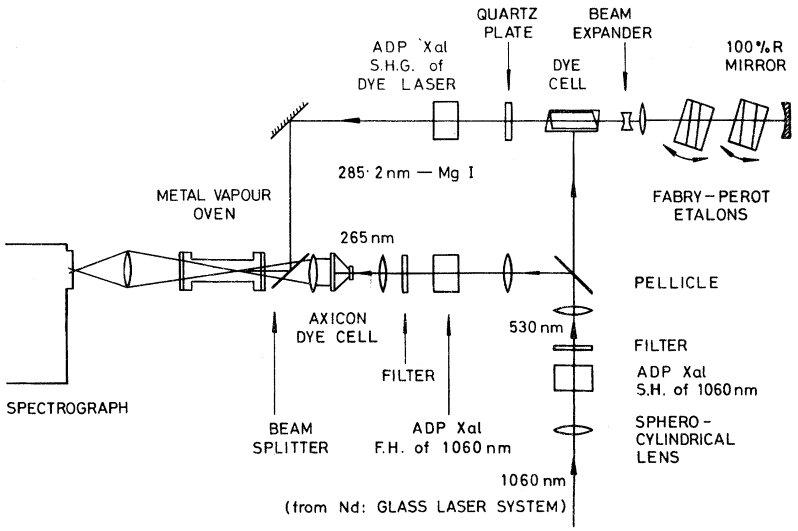

FIG. 1. General experimental arrangement.

tion series, series perturbations, and autoionization in barium[13] have also been investigated experimentally. We have employed these techniques of selective excitation and absorption for the experimental detection and measurement of the calculated $3s3p\,^1P_1^\circ$-$3p^2\,^1S_0$ resonance.

The lifetime of the $3s3p\,^1P_1^\circ$ level is 2 nsec.[14] To record photographically an excited-state absorption of the predicted cross section requires a laser-power density of ~1 MW cm^{-2} at $\lambda = 285.2$ nm.[13] The experimental arrangement employed is shown in Fig. 1. A single-transverse-mode Nd:glass oscillator, actively Q switched with a Pockels cell, was used with amplifiers to produce a 10-J, 30-nsec pulse. Frequency doubling in a phase-matched ammonium dihydrogen phosphate (ADP) crystal generated a 1-J, 22-nsec pulse at $\lambda = 530$ nm. Part of the second harmonic beam (filtered from the fundamental) was used to pump transversely a rhodamine 6-G laser.[15] The dye cell had 10° wedged windows and the 40-cm-long laser resonator was terminated by a 100% reflectivity, concave mirror of 1 m radius of curvature and a quartz parallel plate reflector. Frequency narrowing and tuning with two intracavity Fabry-Perot etalons gave a dye-laser output spectral bandwidth of 0.08 nm in a 22-nsec, 120-mJ pulse, corresponding to a power density of 18 MW cm^{-2} in the 0.3-cm^2 cross-sectional area beam. Beam divergence was 3 mrad, half-angle. Rotation of the two etalons[16] permitted coarse and fine tuning of the dye-laser operating frequency, which was in turn doubled in a second phase-matched ADP crystal to give a 15-mJ, 15-nsec pulse (3.3 MW cm^{-2}), of spectral width 0.04 nm at the $3s^2\,^1S_0$-$3s3p\,^1P_1^\circ$ transition wavelength. 20% of the neodymium second-harmonic beam was reflected by a pellicle beam splitter into another frequency-doubling ADP crystal to generate light at 265 nm. The ultraviolet beam was filtered from the remaining 530-nm light and focused into an axicon dye cell,[13] containing an ultraviolet scintillator dye, to produce a background continuum, with a lifetime of a few nanoseconds,[17] covering the appropriate spectral region. This background continuum light was focused inside the oven.

The metal vapor oven was a simple, resistance-heated, stainless-steel tube of 2 cm internal diameter. Magnesium was kept molten in a reservoir and the 10-cm metal-vapor path length was heated to 650°C. Argon at a pressure of a few hundred torr was used as a buffer gas. A beam splitter, transmitting the fluorescent continuum and reflecting the dye-laser second-harmonic beam, ensured good spatial overlap of the background light with the region of excited atoms in the oven. Coincidence to within 1 nsec of the laser pumping pulse and the background continuum was achieved by adjusting the optical path lengths.

Absorption spectra were recorded with a 1-m Czerny-Turner spectrograph which had a reciprocal dispersion of 0.38 nm/mm in second order. Several laser pulses were required to expose Ilford HP3 or FP4 plates to a density suitable for microdensitometry. To permit accurate comparison of the recorded background continua with

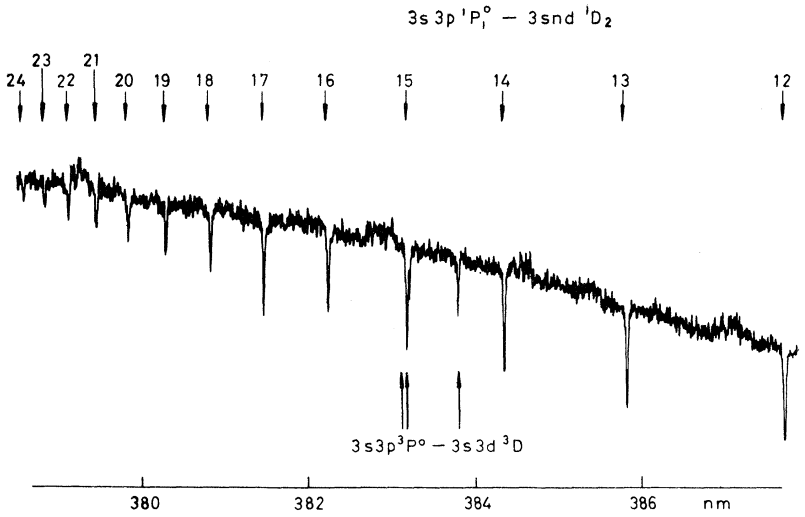

FIG. 2. Microdensitometer trace showing higher terms of the $3s3p\ ^1P_1^\circ$-$3snd\ ^1D_2$ absorption series obtained from selectively excited Mg I. Triplet absorption lines arising from $^1P_1^\circ$-$^3P^\circ$ intersystem crossing are also present.

and without selective excitation of the magnesium vapor, the intensities of the background pulses were monitored with a coaxial photodiode and recorded on a fast oscilloscope.

Absorption spectra were recorded from $\lambda = 400$ to 290 nm. The series $3s3p\ ^1P_1^\circ$-$3snd\ ^1D_2$ was obtained (Fig. 2) in absorption for the first time, up to $n = 24$ (ionization limit at $\lambda = 375.58$ nm), using the spectrograph in second order with a slit width of 50 μm. This extends the series from $n = 13$, previously seen in emission spectra.[18] Figure 3 shows the indole fluorescence continuum, with and without excited magnesium in the oven, recorded in first order with a 200-μm slit width. Strong absorption at $\lambda = 300.9$ nm from the autoionization transition $3s3p\ ^1P_1^\circ$-$3p^2\ ^1S_0$ can be clearly seen near the peak of the continuum. (In this spectral region there are very few suitable fluorescing dyes, and it is not possible to generate a broader spectral background.[17]) There is good agreement with the predicted resonance at $\lambda = 297.7$ nm.[3] By quantitative microdensitometry a linewidth of 3.5 nm [full width at half-maximum (FWHM)] was measured from the absorption-line profile of Fig. 3, obtained by subtracting the normalized background intensity profile. A peak cross section of 5×10^{-16} cm^2 was estimated from the spectral densities and the excited-state population determined by the laser flux. Both of these experimental values are in excellent agreement with those calculated. Figure 3 also shows two sets of triplet emission multiplets. The selec-

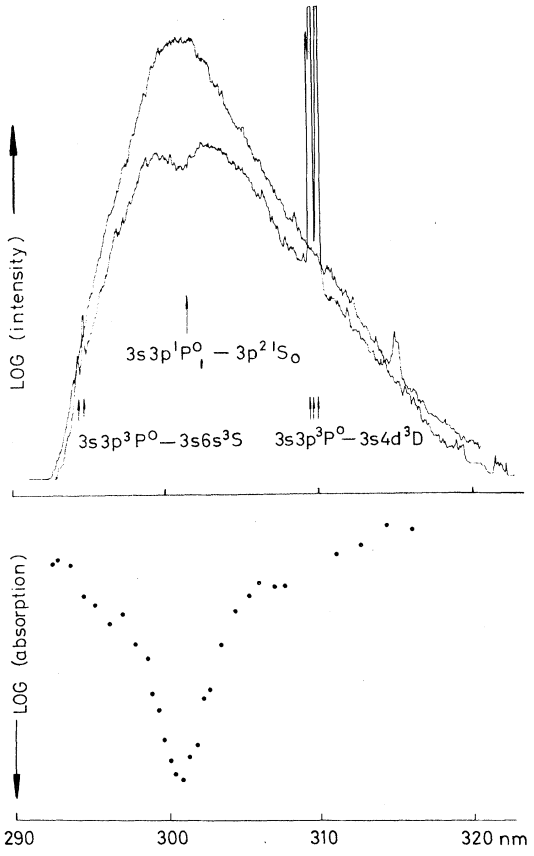

FIG. 3. Top, microdensitometer traces of the indole continuum with and without selective excitation of the magnesium vapor. Bottom, absorption profile of $3s3p\ ^1P_1^\circ$-$3p^2\ ^1S_0$ autoionization line.

tively exciting pump beam at $\lambda = 285.2$ nm will also cause ionization from the $3s3p\ ^1P_1^\circ$ state, and recombination can leave the atom in one of the excited triplet states. We have recorded emission lines terminating in the $3s3p\ ^3P^\circ$ level from $3sns\ ^3S$, $3snd\ ^3D$, and $3p^2\ ^3P$ levels. When the argon buffer gas pressure was increased to 600 Torr from the 300-Torr pressure normally used, these emission lines generally disappeared and often appeared in absorption. This indicates that intersystem crossing is then occurring during the lifetime of the background pulse.

Calculations[3] carried out using a $^1D_2$ continuum state also gave resonance structure in the absorption from the $3s3p\ ^1P_1^\circ$ level at wavelengths less than 200 nm which could be identified as $3pnp\ ^1D$ for $n \geq 4$. Since the present computer code cannot treat satisfactorily the cases of low-energy ejected electrons, the region near to threshold was not investigated. The only conclusion that could be reached about the $3p^2\ ^1D_2$ resonance was that its position is at a wavelength greater than 320 nm. Experiments are in progress to locate this resonance and to measure the $3s3p\ ^1P_1^\circ$-$3p^2\ ^1S_0$ photoionization cross section photoelectrically with a second tunable dye-laser probe.

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Incident X-Ray Energy Dependence of Neon Compton Profiles*

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Exact hydrogenic (EH) Compton-profile calculations have been performed for the L shell of neon. For the scattering of Mo $K\alpha$ and Ag $K\alpha$ radiation from neon the EH calculations show the Mo $K\alpha$ profile to lie higher at the center, in agreement with the experimental results of Eisenberger. An accurate configuration-interaction impulse Compton profile for neon is calculated and it is observed that the effect of correlation of $J(0)$ is significantly smaller than reported earlier.

A technique has recently been developed[1,2] to calculate Compton profiles $J(q)$ within the Born approximation for L-shell electrons, taking the nonrelativistic bound-state and continuum wave functions as hydrogenic in form. The method follows along the lines suggested by Bloch.[3] The wave functions are expressed in parabolic coordinates and certain residue integrations are performed which lead to analytic results[1] for the L-shell matrix elements and Compton profiles. Following the earlier designation of Eisenberger and Platzman[4] who evaluated K-shell Compton