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Two- and three-photon ionization in the noble gases

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By using a characteristic Green's function for an exactly solvable Schrödinger equation with an approximation to the central potential of Hermann and Skillman, the cross section for nonresonant two- and three-photon ionization of Ne, Ar, Kr, and Xe were calculated in *jl* coupling. Expressions for cross sections in *jl* coupling are given. Comparison with the Ar two-photon cross section of Pindzola and Kelly, calculated using the many-body theory, the dipole-length approximation, and *LS* coupling shows a disagreement of as much as a factor of 2. The disagreement appears to arise from distortion introduced by shifting the Green's-function resonances to experimental values.

I. INTRODUCTION

Much of the theoretical and experimental work on atomic multiphoton ionization[1-3] (MPI) has been done on the alkalis. Some experimental work has been published on the alkali earths.[4,5] For the noble gases there have been a number of measurements[6] of ionization due to many (>10) photon absorption, but there are only a small number of papers on ionization in the noble gases due to absorption of a small number of photons.[7-11] This arises from the development of long wavelength glass and ruby laser technology and the high ionization potential of the noble gases compared to the alkalis. However, the rapid development of eximer lasers[12] will allow measurements on noble gas ionization rates due to absorption of two to five photons. For example, the possible NeF laser[13] at 1080 Å can ionize Ne with just two photons. Such systems will be of both scientific and practical significance, allowing the study of Beutler structure,[14] on the one hand, and the possibility of developing reliable nanosecond switches, on the other hand.

The MPI cross sections for the alkalis are dominated by resonances as the energy gap between ground state and low-lying excited levels is small. In this situation intermediate virtual state wave functions can be calculated accurately by summing over a finite number of excited states.[15,16] For the noble gases there is a large gap between the ground and first excited state. Pindzola and Kelly,[8] in calculating the two-photon cross section for Ar, including a sum over both a large discrete set and the continuum, found large differences between their extensive calculation and small basis set approximations which work reasonably well in the alkalis.

Recently I pointed out[17] that by approximating atomic central potentials [actually $rV(r)$] by a series of straight lines, one has a Schrödinger equation exactly solvable in terms of Whittaker functions; but more importantly, one has an exact characteristic Green's function which replaces the infinite sum in the MPI matrix element arising from the eigenfunction expansion of the virtual state. I showed this Green's-function technique produced cross sections for two- and three-photon ionization in the alkalis in reasonable agreement with other calculations and some measurements.

In this paper the characteristic Green's-function technique is used to calculate the two- and three-photon ionization cross section for the noble gases. The calculations are done for both linearly and circularly polarized light, and are done in jl coupling, appropriate for the noble gases.[18] In Sec. II expressions are given for the two- and three-photon ionization cross sections in jl coupling. The one electron eigenvalues occuring with the approximate potential are presented and compared with experimental level energies, and the procedure used to locate resonances at experimental energy levels is described. In Sec. III the cross sections are presented for two-photon ionization and the Ar two-photon cross section is compared with the calculation of Pindzola and Kelly.[8] In Sec. IV the three-photon cross sections are presented.

With the Green's-function technique, the calculation of the radial part of the MPI matrix element is straight forward. The coupling algebra is then the hard part of the calculation. However, the simplicity of the Green's-function technique introduces a difficulty. The Green's function contains one free parameter (the ionization energy). It can be adjusted to produce a resonance at its experimental value. But this choice fixes the energy of all other resonances with the same quantum numbers, and these energies are not, in general, experimental values. In these calculations the free parameter was adjusted in different spectral ranges to produce resonances at experimental values. This led to match-up problems at the boundry of the spectral ranges, i.e., differences of as much as twenty-five percent, and the differences were smoothed over.

II. MECHANICS OF THE CALCULATION

For two-photon ionization the cross sections σ_2^c and σ_2^L, for circularly and linearly polarized light, in units of cm^4 sec are given by

$$\sigma_2^i = 4\pi\left(\frac{\Delta E}{F_0}\right)^2 (M_2^i)^2 , \tag{1}$$

where ΔE is the photon energy in Ry, the continuum orbital is normalized per Ry, the energy in the Green's function is in Ry,[19] and $F_0 = 3.22\times10^{34}/\text{cm}^2\,\text{sec}$. The intermediate state is described by the quantum numbers $J_f, n_1 l_1 K_1 J_1$, where J_f ($\frac{3}{2}$ or $\frac{1}{2}$) is the total angular momentum of the $(np)^5$ core of the noble gas ion, and $\vec{K}_1 = \vec{J}_f + \vec{l}_1$ and $\vec{K}_1 + \vec{\frac{1}{2}} = \vec{J}_1$ defines jl coupling. Since J_1 must be 1, the energy of the intermediate state is defined by $E_1(J_f, n_1 l_1 K_1\, 1)$. In terms of the quantities[20]

$$P(J_f, 1) = \sum_{n_1} \frac{\langle n_0 p | r | n_1 s\rangle\langle n_1 s | r | \epsilon p\rangle}{[E_1(J_f, n_1 s \tfrac{3}{2} 1) - E_0 - \omega]} , \tag{2a}$$

$$Q(J_f, 1) = \sum_{n_1} \frac{\langle n_0 p | r | n_1 s\rangle\langle n_1 s | r | \epsilon p\rangle}{[E_1(J_f, n_1 s \tfrac{1}{2} 1) - E_0 - \omega]} , \tag{2b}$$

$$S(J_f, l_f) = \sum_{n_1} \frac{\langle n_0 p | r | n_1 d\rangle\langle n_1 d | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 d \tfrac{3}{2} 1) - E_0 - \omega]} , \tag{2c}$$

$$T(J_f, l_f) = \sum_{n_1} \frac{\langle n_0 p | r | n_1 d\rangle\langle n_1 d | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 d \tfrac{1}{2} 1) - E_0 - \omega]} , \tag{2d}$$

$$\begin{aligned}(M_2^c)^2 = {} & \tfrac{2}{15}\left|T(\tfrac{3}{2}, 3) + \tfrac{1}{5} S(\tfrac{3}{2}, 3)\right|^2 + \tfrac{16}{125} S(\tfrac{3}{2}, 3)^2 + \tfrac{4}{25} S(\tfrac{1}{2}, 3)^2 + \tfrac{2}{15}\left|P(\tfrac{3}{2}, 1) + \tfrac{1}{5} S(\tfrac{3}{2}, 1)\right|^2 \\ & + \tfrac{2}{135}\left|P(\tfrac{3}{2}, 1) - \tfrac{4}{5} S(\tfrac{3}{2}, 1) + T(\tfrac{3}{2}, 1)\right|^2 + \tfrac{2}{27}\left|Q(\tfrac{1}{2}, 1) + \tfrac{1}{5} S(\tfrac{1}{2}, 1)\right|^2\end{aligned} \tag{3a}$$

and

$$(M_2^L)^2 = \tfrac{2}{3}(M_2^C)^2 + \tfrac{4}{81}\left|P(\tfrac{3}{2}, 1) + S(\tfrac{3}{2}, 1) + T(\tfrac{3}{2}, 1)\right|^2 + \tfrac{2}{81}\left|Q(\tfrac{1}{2}, 1) + 2S(\tfrac{1}{2}, 1)\right|^2 . \tag{3b}$$

In terms of the radial Green's function defined in Ref. 17,

$$\sum_{n_1} \frac{\langle n_0 p | r | n_1 l_1\rangle\langle n_1 l_1 | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 l_1 K_1, 1) - E_0 - \omega]} = \int_0^\infty r_1^3 dr_1 \Psi_{n_0 p}(r_1) \int_0^\infty r_2^3 dr_2 \Psi_{\epsilon l f}(r_2) g_{l_1}(r_1, r_2, E_0 + \omega) . \tag{4}$$

For the matrix element to diverge at the experimental $E_1(J_f, N_1 l_1 K_1 1)$, E_0 is changed to $E_0 - E(n_1 l_1) + E_1(J_f, n_1 l_1 K_1 1)$, where $E(n_1 l_1)$ is the model one electron eigenvalue. For a particular range in ω it is assumed that only the nearest resonance with a particular l value need have its resonance energy specified exactly. By changing E_0, one shifts all the resonances with a given J_f, l_1 and K_1. For the two-photon case, eight Green's functions are calculated. The energy of the continuum orbital is either $2\omega - E_0$ or $2\omega - E_0 - \Delta_{so}$, where E_0 is the atomic ionization energy and Δ_{so} is the spin-orbit splitting in the ionic ground state.

For three-photon ionization the cross section, in cm^6 sec^2

$$\sigma_3^i = 8\pi(\Delta E/F_0)^3 (M_c^i)^2 , \tag{5}$$

with

$$P(J_f, K_1, K_2 J_2, l_f) = \sum_{n_1 n_2} \frac{\langle n_0 p | r | n_1 s\rangle\langle n_1 s | r | n_2 p\rangle\langle n_2 p | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 s K_1, 1) - E_0 - \omega][E_2(J_f, n_2 p K_2, J_2) - E_0 - 2\omega]} , \tag{6a}$$

$$Q(J_j, K_1, K_2 J_2, l_f) = \sum_{n_1 n_2} \frac{\langle n_0 p | r | n_1 d\rangle\langle n_1 d | r | n_2 f\rangle\langle n_2 f | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 d K_1, 1) - E_0 - \omega][E_2(J_f, n_2 f K_2, J_2) - E_0 - 2\omega]} , \tag{6b}$$

$$S(J_f, K_1, K_2 J_2, l_f) = \sum_{n_1 n_2} \frac{\langle n_0 p | r | n_1 d\rangle\langle n_1 d | r | n_2 p\rangle\langle n_2 p | r | \epsilon l_f\rangle}{[E_1(J_f, n_1 d K_1, 1) - E_0 - \omega][E_2(J_f, n_2 p K_2, J_2) - E_0 - 2\omega]} , \tag{6c}$$

then

$$\begin{aligned}(M_3^c)^2 = {} & \tfrac{16}{315}\left|Q(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\, 2, 4) + \tfrac{1}{5} Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\, 2, 4) + \tfrac{3}{35} Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 4)\right|^2 + \tfrac{16}{343} Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 4)^2 + \tfrac{16}{245} Q(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 4)^2 \\ & + \tfrac{16}{315}\left|P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2) + \tfrac{1}{5} S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2) + \tfrac{3}{35} Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2)\right|^2 \\ & + \tfrac{8}{945}\left|\tfrac{1}{10}[3P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2) + 7P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\, 2, 2)] + \tfrac{1}{50}[3S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2) - 28S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2} 2, 2) + 35S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2} 2, 2]\right. \\ & \left. - \tfrac{1}{350}[96Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2) - 21Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\, 2, 2) - 105Q(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\, 2, 2)]\right|^2 + \tfrac{4}{135}\left|P(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{3}{2}\, 2, 2) + \tfrac{1}{5} S(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{3}{2}\, 2, 2)\right. \\ & \left. + \tfrac{3}{35} Q(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{5}{2}\, 2, 2)\right|^2 ,\end{aligned} \tag{7a}$$

and

$$
\begin{aligned}
(M_3^L)^2 = {} & \tfrac{2}{5}(M_3^c)^2 + \tfrac{4}{729}\,\big|P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 0) + S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 0) + S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{1}{2}\; 0, 0) + \tfrac{1}{5}[9P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 0) + P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 0)] \\
& + \tfrac{1}{25}[9S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 0) - 4S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 0) + 5S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 0)]\big|^2 \\
& + \tfrac{2}{729}\,\big|P(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\; 0, 0) + 2S(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 0) + 2P(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{3}{2} 2, 0) + \tfrac{2}{5}S(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 0)\big|^2 \\
& + \tfrac{4}{729}\,\big|P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 2) + S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 2) + S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{1}{2}\; 0, 2) + \tfrac{1}{25}[9P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 2) - 4P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2)] \\
& \qquad + \tfrac{1}{125}[9S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 2) + 16S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2) - 20S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 2)] \\
& \qquad + \tfrac{9}{125}[24Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 2) + Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2) + 5Q(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 2)]\big|^2 \\
& + \tfrac{4}{729}\,\big|P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 2) + S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 2) + S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{1}{2}\; 0, 2) + \tfrac{1}{5}P(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2) + \tfrac{1}{25}[5S(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 2) - 4S(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2)] \\
& \qquad + \tfrac{9}{25}[Q(\tfrac{3}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2) + 5Q(\tfrac{3}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 2)]\big|^2 + \tfrac{4}{729}\,\big|P(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\; 0, 2) + 2S(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{1}{2}\; 0, 2) + \tfrac{54}{25}Q(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{5}{2}\; 2, 2) \\
& \qquad\qquad + \tfrac{1}{25}S(\tfrac{1}{2}, \tfrac{3}{2}, \tfrac{3}{2}\; 2, 2) + \tfrac{1}{5}P(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{3}{2}\; 2, 2)\big|^2 .
\end{aligned}
\tag{7b}
$$

In terms of the radial Green's functions defined in Ref. 17

$$
\begin{aligned}
& \sum_{n_1, n_2} \frac{\langle n_0 p | r | n_1 l_1 \rangle \langle n_1 l_1 | r | n_2 l_2 \rangle \langle n_2 l_2 | r | \epsilon l_f \rangle}{[E_1(J_f, n_1 l_1 K_1, 10) - E_0 - \omega][E_2(J_f, n_2 l_2 K_2, J_2) - E_0 - 2\omega]} \\
& \quad = \int_0^\infty r_1^3 dr_1 \Psi_{n_0 p}(r_1) \int_0^\infty r_2^3 dr_2 g_{l_1}(r_1, r_2, E_0 + \omega) \int_0^\infty r_3^3 dr_3 \Psi_{\epsilon l_f}(r_3) g_{l_2}(r_2, r_3, E_0 + 2\omega) .
\end{aligned}
\tag{8}
$$

In the calculation it was assumed that g_{l_1} was independent of K_1 and four Green's functions were used corresponding to $J_f = \frac{1}{2}, \frac{3}{2}$ and $l_1 = s, d$. For the second term, eight Green's functions were used, i.e., $l_2 = 3$, $J_f K_2 J_2 = (\frac{3}{2}, \frac{3}{2}, 2)$, $(\frac{3}{2}, \frac{5}{2}, 2)$, and $(\frac{1}{2}, \frac{5}{2}, 2)$, and for $l_f = 1, J_f K_2 J_2 = (\frac{3}{2}, \frac{5}{2}, 2)$, $(\frac{3}{2}, \frac{3}{2}, 2)$, $(\frac{3}{2}, \frac{1}{2}, 0)$, $(\frac{1}{2}, \frac{3}{2}, 2)$, and $(\frac{1}{2}, \frac{1}{2}, 0)$. As in the two-photon case we adjust E_0 in g_{l_2} so that the resonances in the three-photon cross section occur at the experimental energy levels.

Since an adjustment in E_0 in each of the Green's functions shifts all the resonances with the same l, K, J values, the adjustment is reasonably accurate only if the adjustment is considerably smaller than the level separation. The level designation used, and experimental and calculated energies and adjustments for Ne, Ar, Kr, and Xe, are listed in Tables I–IV, respectively. Generally the adjustments are less than ten percent of the separation of corresponding levels, though there are exceptions. Levels, not assigned in Moore's tables,[18] are omitted.

III. TWO-PHOTON IONIZATION CROSS SECTIONS

The calculated cross sections for Ne–Xe are shown in Figs. 1–4, respectively. The figures show the cross section for circularly polarized light and the ratio of circular cross section to linear cross section. The ratio $R = \sigma_2^C / \sigma_2^L$ has a narrow range of variation in the noble gases. From Eqs. (3a) and (3b), when the matrix elements to continuum f orbitals dominate the matrix elements to continuum p orbitals, $R = \frac{3}{2}$. At $(np)^5\ (n's)$ resonances, $R = 1$. At $(np)^5\ {}^2P_{3/2}$, $(n'd)_{k=3/2}$ resonances (designated *nd* in Tables I–IV and in the figures),

$$R = \tfrac{3}{2}[S(\tfrac{3}{2}, 3)^2 + \tfrac{1}{9}S(\tfrac{3}{2}, 1)^2] / [S(\tfrac{3}{2}, 3)^2 + \tfrac{2}{3}S(\tfrac{3}{2}, 1)^2] .$$

If the $l_f = 3$ matrix element is dominant $R = \frac{3}{2}$ and if the $l_f = 1$ matrix element is dominant $R = \frac{1}{4}$. At $(np)^5\ {}^2P_{1/2}$, $(n'd)_{k=3/2}$ resonances (designated *nd″* in Tables I–IV and in the figures),

$$R = \tfrac{3}{2}[S(\tfrac{1}{2}, 3)^2 + \tfrac{1}{54}S(\tfrac{1}{2}, 1)^2] / [S(\tfrac{1}{2}, 3)^2 + \tfrac{17}{18}S(\tfrac{1}{2}, 1)^2] .$$

If $S(\frac{1}{2}, 1)^2 \gg S(\frac{1}{2}, 3)^2$, R can be as small as $\frac{1}{34}$. At $(np)^5\ {}^2P_{3/2}$, $(n'd)_{k=1/2}$ resonances (designated *nd′* in Tables I–IV and the figures),

$$R = \tfrac{3}{2}[T(\tfrac{3}{2}, 3)^2 + \tfrac{1}{9}T(\tfrac{3}{2}, 1)^2] / [T(\tfrac{3}{2}, 3)^2 + \tfrac{2}{3}T(\tfrac{3}{2}, 1)^2] .$$

R can be as low as $\frac{1}{4}$ if $T(\frac{3}{2}, 1)^2 \gg T(\frac{3}{2}, 3)^2$.

The structure in R described above is due to competition between continuum final states. Addition structure can arise from interference between *nd* and *nd′* resonances providing the resonances are close together, so the matrix elements are large enough to dominate the background, and the matrix elements have opposite sign. The only instance where significant structure in R appeared in the two-photon cross sections was the 3*d*, 3*d′*, 3*d″* resonance in Ne.

The wavelength scale in the figures is not linear; it is compressed when the cross sections vary slowly and expanded near resonances. In Fig. 1 for Ne at 1 Å per division near the 3*d* resonances, the 3*d* and 3*d′* resonances overlap. The 3*d″* resonance is merely a glitch in the curve. The dashed curve in Fig. 1 is 3*d″* resonance on a scale of 0.02 Å/division. But on a 1 Å/division scale the struc-

TABLE I. Level designations, quantum numbers, and experimental and calculated energies and differences (in cm^{-1}) relative to the $(2p)^5\,{}^2P_{3/2}$ and ${}^2P_{1/2}$ ion levels for Ne.

$J_f=\frac{3}{2}$					$J_f=\frac{1}{2}$				
desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})	desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})
$3s$	$\frac{3}{2},1$	39 471	36 698	2723	$3s'$	$\frac{1}{2},1$	38 821	36 698	2123
$4s$	$\frac{3}{2},1$	15 134	14 613	521	$4s'$	$\frac{1}{2},1$	15 175	14 613	562
$5s$	$\frac{3}{2},1$	8017	7829	188	$5s'$	$\frac{1}{2},1$	8054	7829	225
$6s$	$\frac{3}{2},1$	4963	4872	91	$6s'$	$\frac{1}{2},1$	4982	4872	110
$7s$	$\frac{3}{2},1$	3373	3323	50	$7s'$	$\frac{1}{2},1$	3386	3323	63
$3d$	$\frac{3}{2},1$	12 293	12 187	106	$3d''$	$\frac{3}{2},1$	12 274	12 187	85
$3d'$	$\frac{1}{2},1$	12 406		219					
$4d$	$\frac{3}{2},1$	6903	6855	48	$4d''$	$\frac{3}{2},1$	6902	6855	47
$4d'$	$\frac{1}{2},1$	6962		107					
$5d$	$\frac{3}{2},1$	4413	4387	26	$5d''$	$\frac{3}{2},1$	4414	4387	27
$5d'$	$\frac{1}{2},1$	4442		55					
$6d$	$\frac{3}{2},1$	3062	3047	15	$6d''$	$\frac{3}{2},1$	3065	3047	18
$6d'$	$\frac{1}{2},1$	3079		32					
$7d$	$\frac{3}{2},1$	2247	2238	9	$7d''$	$\frac{3}{2},1$	2249	2238	9
$7d'$	$\frac{1}{2},1$	2258		20					
$3p$	$\frac{5}{2},2$	24 106		1175	$3p'''$	$\frac{3}{2},2$	23 852	22 931	921
$3p'$	$\frac{3}{2},2$	23 615	22 931	684	$3p''''$	$\frac{1}{2},0$	21 739		−1192
$3p''$	$\frac{1}{2},0$	23 013		82					
$4p$	$\frac{5}{2},2$	11 031		340	$4p'''$	$\frac{3}{2},2$	11 001	10 691	310
$4p'$	$\frac{3}{2},2$	10 892	10 691	201	$4p''''$	$\frac{1}{2},0$	10 424		−267
$4p''$	$\frac{1}{2},0$	10 529		−162					
$5p$	$\frac{5}{2},2$	6339		146	$5p'''$	$\frac{3}{2},2$	6331	6193	138
$5p'$	$\frac{3}{2},2$	6281	6193	88	$5p''''$	$\frac{1}{2},0$	6123		−70
$5p''$	$\frac{1}{2},0$	6063		−130					
$6p$	$\frac{5}{2},2$	4115		76	$6p'''$	$\frac{3}{2},2$	4113	4039	74
$6p'$	$\frac{3}{2},2$	4086	4039	47	$6p''''$	$\frac{1}{2},0$	4021		−18
$6p''$	$\frac{1}{2},0$	3953		−86					
$7p$	$\frac{5}{2},2$	2886		44	$7p'''$	$\frac{3}{2},2$	2882	2842	40
$7p'$	$\frac{3}{2},2$	2870	2842	28	$7p''''$	$\frac{1}{2},0$	2797		−45
$7p''$	$\frac{1}{2},0$	2781		−61					
$4f$	$\frac{5}{2},2$	6861	6855	6	$4f''$	$\frac{5}{2},2$	6863	6855	8
$4f'$	$\frac{3}{2},2$	6877		22					
$5f$	$\frac{5}{2},2$	4391	4387	4	$5f''$	$\frac{5}{2},2$	4392	4387	5
$5f'$	$\frac{3}{2},2$	4400		13					

TABLE II. Level designations, quantum numbers, and experimental and calculated energies and differences (in cm^{-1}) relative to the $(3p)^5\,{}^2P_{3/2}$ and ${}^2P_{1/2}$ ion levels for Ar.

		$J_f=\frac{3}{2}$					$J_f=\frac{1}{2}$		
desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})	desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})
$4s$	$\frac{3}{2}$, 1	33 359	30 316	3043	$4s'$	$\frac{1}{2}$, 1	33 141	30 316	2825
$5s$	$\frac{3}{2}$, 1	13 467	12 885	582	$5s'$	$\frac{1}{2}$, 1	13 566	12 885	681
$6s$	$\frac{3}{2}$, 1	7350	7129	221	$6s'$	$\frac{1}{2}$, 1	7380	7129	251
$7s$	$\frac{3}{2}$, 1	4361	4522	109	$7s'$	$\frac{1}{2}$, 1	4659	4522	137
$8s$	$\frac{3}{2}$, 1	3174	3123	51	$8s'$	$\frac{1}{2}$, 1	3188	3123	65
$3d$	$\frac{3}{2}$, 1	12 962	12 540	422	$3d''$	$\frac{3}{2}$, 1	13 174	12 540	634
$3d'$	$\frac{1}{2}$, 1	15 292		2752					
$4d$	$\frac{3}{2}$, 1	7262	7083	179	$4d''$	$\frac{3}{2}$, 1	7529	7082	446
$4d'$	$\frac{1}{2}$, 1	8459		1376					
$5d$	$\frac{3}{2}$, 1	4596	4525	71	$5d''$	$\frac{3}{2}$, 1	4725	4525	200
$5d'$	$\frac{1}{2}$, 1	5177		652					
$6d$	$\frac{3}{2}$, 1		3134		$6d''$	$\frac{3}{2}$, 1	3255	3134	121
$6d'$	$\frac{1}{2}$, 1	3642		508					
$7d$	$\frac{3}{2}$, 1	2322			$7d''$	$\frac{3}{2}$, 1			
$7d'$	$\frac{1}{2}$, 1	2555							
$4p$	$\frac{5}{2}$, 2	21 493		1503	$4p'''$	$\frac{3}{2}$, 2	21 251	19 990	1261
$4p'$	$\frac{3}{2}$, 2	20 872	19 990	882	$4p''''$	$\frac{1}{2}$, 0	19 818		−172
$4p''$	$\frac{1}{2}$, 0	20 056		66					
$5p$	$\frac{5}{2}$, 2	10 111		435	$5p'''$	$\frac{3}{2}$, 2	10 072	9676	396
$5p'$	$\frac{3}{2}$, 2	9926	9676	250	$5p''''$	$\frac{1}{2}$, 0	9670		−6
$5p''$	$\frac{1}{2}$, 0	9547		−129					
$6p$	$\frac{5}{2}$, 2	5918		189	$6p'''$	$\frac{3}{2}$, 2	5906	5729	177
$6p'$	$\frac{3}{2}$, 2	5839	5729	110	$6p''''$	$\frac{1}{2}$, 0	5750		21
$6p''$	$\frac{1}{2}$, 0	5640		−89					
$7p$	$\frac{5}{2}$, 2	3889		99	$7p'''$	$\frac{3}{2}$, 2	3882	3790	92
$7p'$	$\frac{3}{2}$, 2	3848	3790	58	$7p''''$	$\frac{1}{2}$, 0	3791		1
$7p''$	$\frac{1}{2}$, 0	3725		−65					
$8p$	$\frac{5}{2}$, 2	2753			$8p'''$	$\frac{3}{2}$, 2	2749		
$8p'$	$\frac{3}{2}$, 2	2729			$8p''''$	$\frac{1}{2}$, 0	2710		
$8p''$	$\frac{1}{2}$, 0	2671							
$4f$	$\frac{5}{2}$, 2	6880	6855	25	$4f''$	$\frac{5}{2}$, 2	6886	6855	31
$4f'$	$\frac{3}{2}$, 2	6921		66					
$5f$	$\frac{5}{2}$, 2	4402	4387	15	$5f''$	$\frac{5}{2}$, 2	4404	4387	17
$5f'$	$\frac{3}{2}$, 2	4424		37					

TABLE III. Level designations, quantum numbers, and experimental and calculated energies and differences (in cm^{-1}) relative to the $(4p)^5\,{}^2P_{3/2}$ and ${}^2P_{1/2}$ ion levels for Kr.

		$J_f=\frac{3}{2}$					$J_f=\frac{1}{2}$		
desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})	desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})
$5s$	$\frac{3}{2},1$	31 997	28 516	3481	$5s'$	$\frac{1}{2},1$	32 437	28 516	3921
$6s$	$\frac{3}{2},1$	13 020	12 373	647	$6s'$	$\frac{1}{2},1$	13 138	12 373	765
$7s$	$\frac{3}{2},1$	7143	6915	228	$7s'$	$\frac{1}{2},1$	7281	6915	366
$8s$	$\frac{3}{2},1$	4541	4414	127	$8s'$	$\frac{1}{2},1$	4574	4414	160
$9s$	$\frac{3}{2},1$	3135	3061	74	$9s'$	$\frac{1}{2},1$	3157	3061	96
$4d$	$\frac{3}{2},1$	13 268	13 007	261	$4d''$	$\frac{3}{2},1$	13 397	13 007	390
$4d'$	$\frac{1}{2},1$	15 829		2822					
$5d$	$\frac{3}{2},1$	7266	7368	−102	$5d''$	$\frac{3}{2},1$	7551	7368	183
$5d'$	$\frac{1}{2},1$	9112		1744					
$6d$	$\frac{3}{2},1$	4655	4689	−34	$6d''$	$\frac{3}{2},1$	4785	4689	96
$6d'$	$\frac{1}{2},1$	5238		549					
$7d$	$\frac{3}{2},1$	3225	3233	−8	$7d''$	$\frac{3}{2},1$	3275	3233	42
$7d'$	$\frac{1}{2},1$	3571		338					
$8d$	$\frac{3}{2},1$	2400			$8d''$	$\frac{3}{2},1$	2375		
$8d'$	$\frac{1}{2},1$	2624							
$5p$	$\frac{5}{2},2$	20 607		1774	$5p'''$	$\frac{3}{2},2$	20 339	18 829	1510
$5p'$	$\frac{3}{2},2$	19 791	18 829	962	$5p''''$	$\frac{1}{2},0$	19 429		600
$5p''$	$\frac{1}{2},0$	18 820		−9					
$6p$	$\frac{5}{2},2$	9793		530	$6p'''$	$\frac{3}{2},2$	9716	9263	453
$6p'$	$\frac{3}{2},2$	9552	9263	289	$6p''''$	$\frac{1}{2},0$	9463		200
$6p''$	$\frac{1}{2},0$	9153		−110					
$7p$	$\frac{5}{2},2$	5773		236	$7p'''$	$\frac{3}{2},2$			
$7p'$	$\frac{3}{2},2$	5668	5537	131	$7p''''$	$\frac{1}{2},0$			
$7p''$	$\frac{1}{2},0$	5504		−33					
$8p$	$\frac{5}{2},2$	3808		123	$8p'''$	$\frac{3}{2},2$			
$8p'$	$\frac{3}{2},2$	3753	3685	68	$8p''''$	$\frac{1}{2},0$			
$8p''$	$\frac{1}{2},0$	3618		−67					
$9p$	$\frac{5}{2},2$	2704		74					
$9p'$	$\frac{3}{2},2$	2671	2630	41					
$9p''$	$\frac{1}{2},0$	2606		−24					
$4f$	$\frac{5}{2},2$	6893	6855	38	$4f''$	$\frac{5}{2},2$	6903	6855	48
$4f'$	$\frac{3}{2},2$	6949		94					
$5f$	$\frac{5}{2},2$	4410	4387	23	$5f''$	$\frac{5}{2},2$			
$5f'$	$\frac{3}{2},2$	4443		56					

TABLE IV. Level designations, quantum numbers, and experimental and calculated energies and differences (in cm^{-1}) relative to the $(5p)^5\,{}^2P_{3/2}$ and ${}^2P_{1/2}$ ion levels for Xe.

$J_f=\frac{3}{2}$					$J_f=\frac{1}{2}$				
desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})	desig.	K,J	E (cm^{-1})	E_{CALC}	Δ (cm^{-1})
$6s$	$\frac{3}{2},1$	29 788	24 961	4827	$6s'$	$\frac{1}{2},1$	31 185	24 961	6224
$7s$	$\frac{3}{2},1$	12 393	11 293	1100	$7s'$	$\frac{1}{2},1$	12 570	11 293	1277
$8s$	$\frac{3}{2},1$	6901	6451	450	$8s'$	$\frac{1}{2},1$	6945	6451	494
$9s$	$\frac{3}{2},1$	4411	4173	238	$9s'$	$\frac{1}{2},1$	4417	4173	244
$10s$	$\frac{3}{2},1$	3046	2921	125	$10s'$	$\frac{1}{2},1$	3055	2921	134
$5d$	$\frac{3}{2},1$	13 944	13 173	771	$5d''$	$\frac{3}{2},1$	14 752	13 173	1579
$5d'$	$\frac{1}{2},1$	17 847		4674					
$6d$	$\frac{3}{2},1$	7801	7450	351	$6d''$	$\frac{3}{2},1$	7953	7450	503
$6d'$	$\frac{1}{2},1$	9284		1834					
$7d$	$\frac{3}{2},1$	5119	4730	389	$7d''$	$\frac{3}{2},1$	4952	4730	222
$7d'$	$\frac{1}{2},1$	5705		975					
$8d$	$\frac{3}{2},1$	3148	3255	−107	$8d''$	$\frac{3}{2},1$	3342	3255	87
$8d'$	$\frac{1}{2},1$	3605		350					
$9d$	$\frac{3}{2},1$	2335	2373	−38	$9d''$	$\frac{3}{2},1$	2406	2373	33
$9d'$	$\frac{1}{2},1$	2605		232					
$6p$	$\frac{5}{2},2$	19 714		2309	$6p'''$	$\frac{3}{2},2$	19 208	17 405	1803
$6p'$	$\frac{3}{2},2$	18 621	17 405	1216	$6p''''$	$\frac{1}{2},0$	18 510		1105
$6p''$	$\frac{1}{2},0$	17 715		310					
$7p$	$\frac{5}{2},2$	9482		740	$7p'''$	$\frac{3}{2},2$			
$7p'$	$\frac{3}{2},2$	9147	8742	705	$7p''''$	$\frac{1}{2},0$			
$7p''$	$\frac{1}{2},0$	8991		249					
$8p$	$\frac{5}{2},2$	5612		320	$8p'''$	$\frac{3}{2},2$			
$8p'$	$\frac{3}{2},2$	5463	5292	171	$8p''''$	$\frac{1}{2},0$			
$8p''$	$\frac{1}{2},0$	5278		−14					
$9p$	$\frac{5}{2},2$	3723		173	$9p'''$	$\frac{3}{2},2$			
$9p'$	$\frac{3}{2},2$	3643	3550	93	$9p''''$	$\frac{1}{2},0$			
$9p''$	$\frac{1}{2},0$	3548		−2					
$10p$	$\frac{5}{2},2$	2652		104	$10p'''$	$\frac{3}{2},2$			
$10p'$	$\frac{3}{2},2$	2604	2548	56	$10p''''$	$\frac{1}{2},0$			
$10p''$	$\frac{1}{2},0$	2547		−1					
$4f$	$\frac{5}{2},2$	6923	6855	68	$7f''$	$\frac{5}{2},2$			
$4f'$	$\frac{3}{2},2$	6984		129					
$5f$	$\frac{5}{2},2$	4430	4387	43	$5f''$	$\frac{5}{2},2$			
$5f'$	$\frac{3}{2},2$	4467		80					

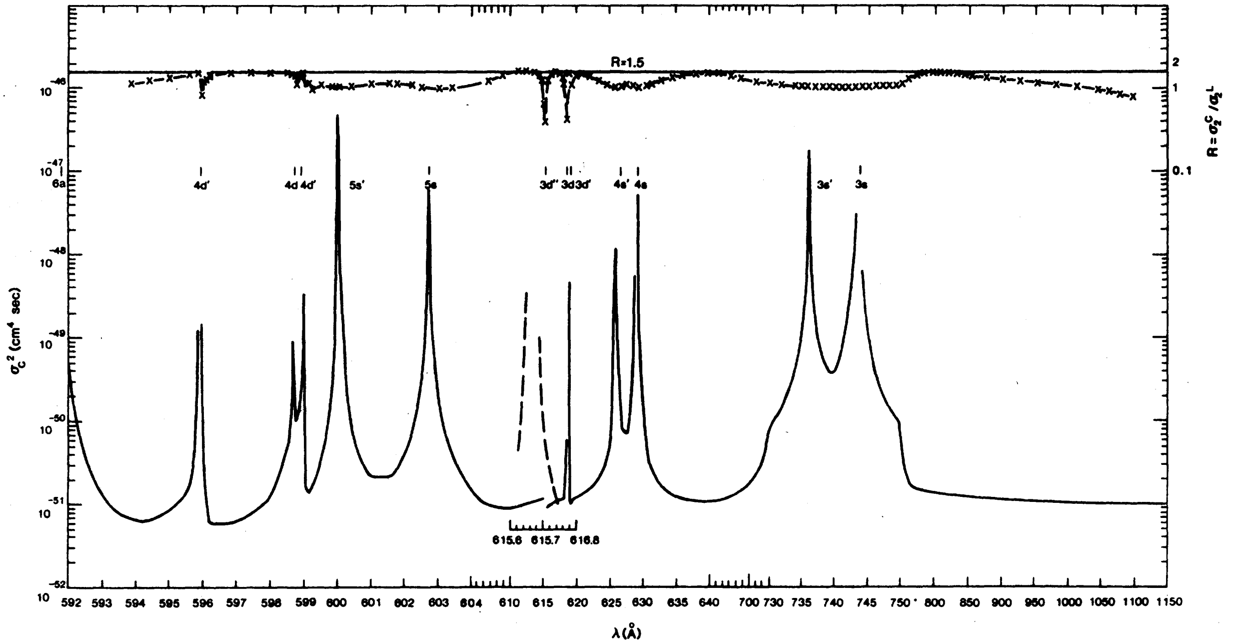

FIG. 1. The cross section for two-photon ionization with circularly polarized light and $R=\sigma_2^C/\sigma_2^L$ for Ne. The wavelength scale has been expanded and contracted to show resonance structure. The dashed curve and scale of 0.02 Å/division illustrates the narrowness of the $3d''$ resonance.

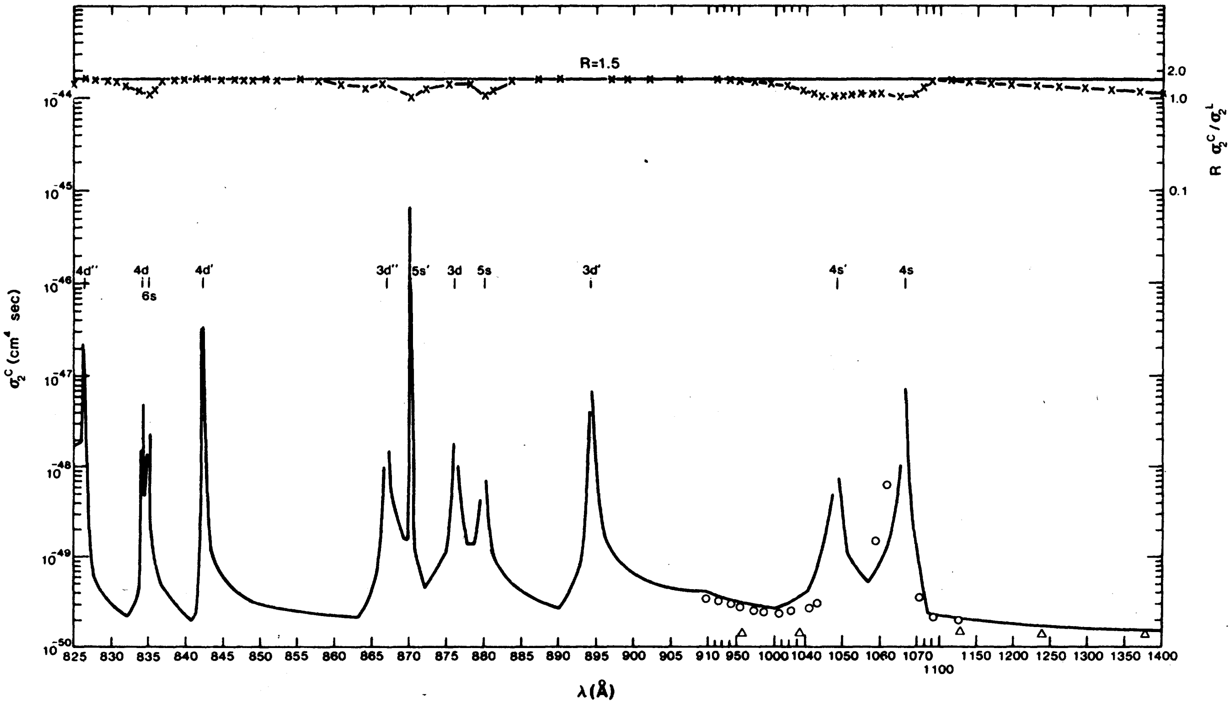

FIG. 2. Same as Fig. 1 for Ar. The open triangles are from the *LS*-coupling calculations of Ref. 8, while the open circles are my *LS*-coupling calculations.

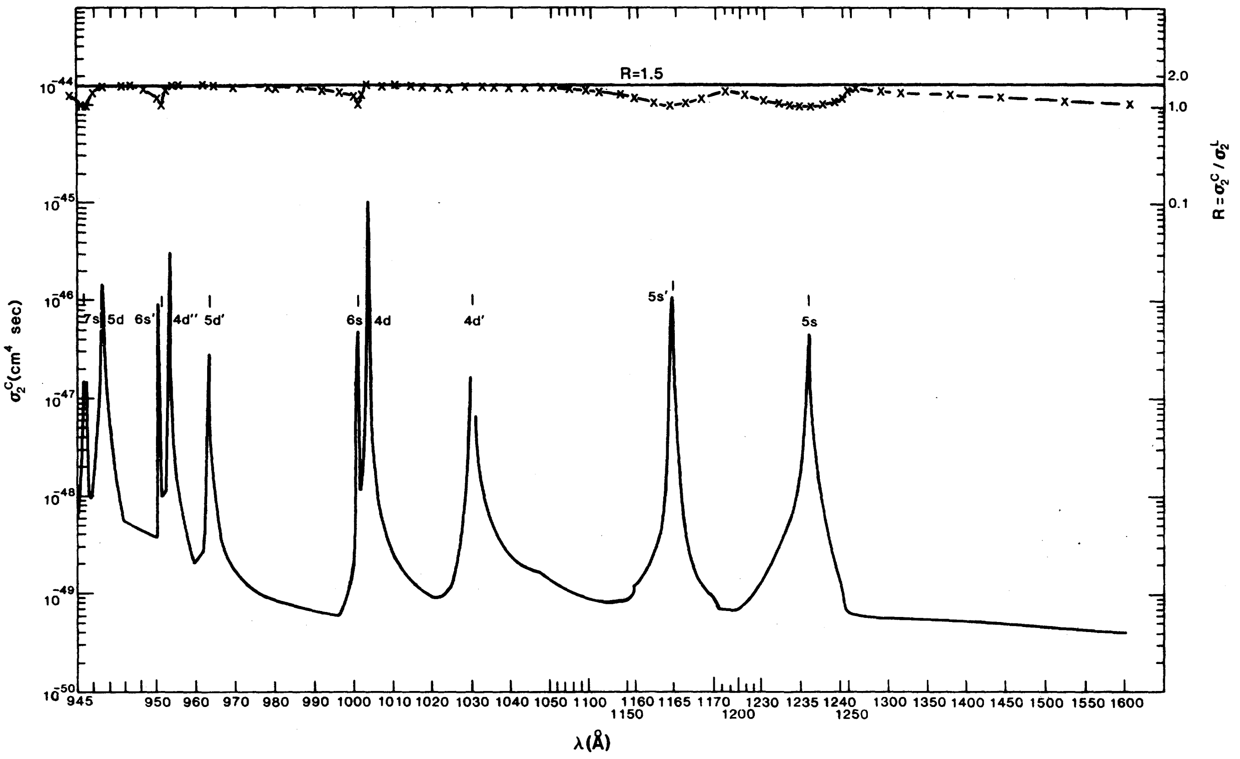

FIG. 3. Same as Fig. 1 for Kr.

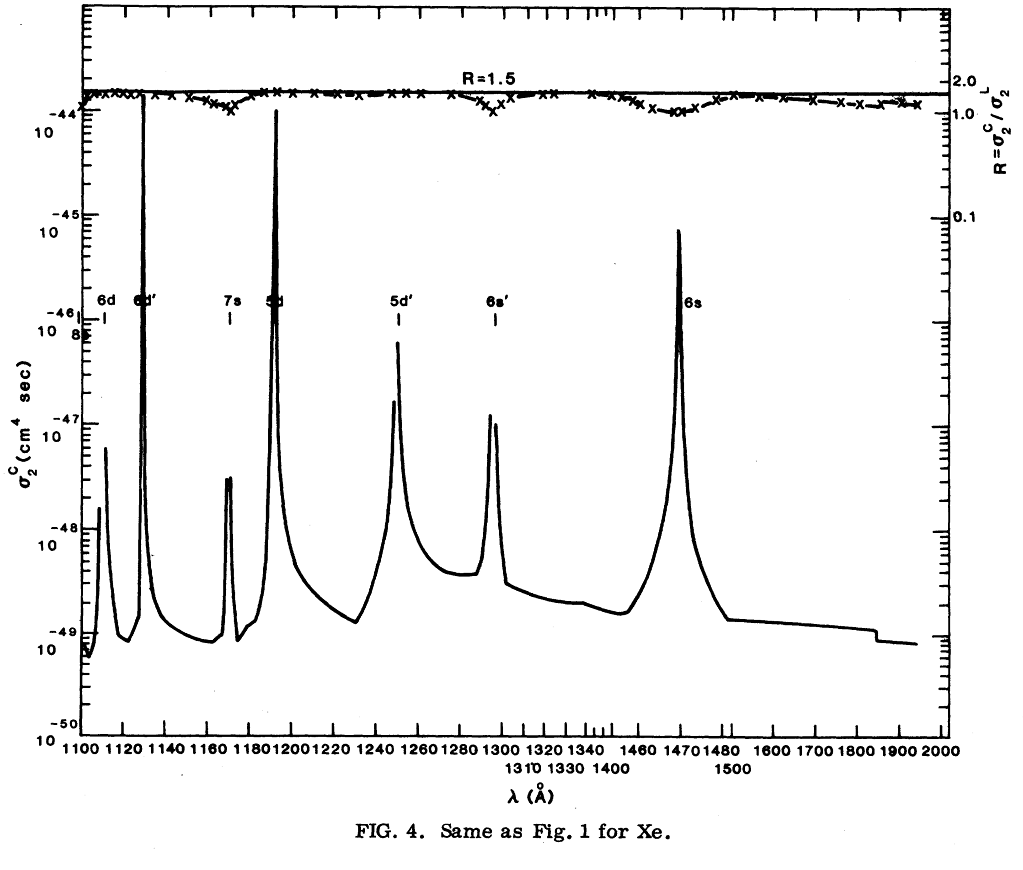

FIG. 4. Same as Fig. 1 for Xe.

ture in R is apparent. The dip in R at the $3d''$ resonance is due to the dominance of the $l_f=1$ channel compared to the $l_f=3$ channel. The dip-peak-dip structure in R at the $3d$–$3d'$ resonances arises from dominance of the $l_f=1$ channel (2 dips), and the interference between the resonances of the $l_f=1$ channel leading to dominance of the $l_f=3$ matrix elements (peak).

In Fig. 2 for Ar some points from the calculations of Pindzola and Kelly are shown as open triangles. Pindzola and Kelly[8] present values for σ_2^L, and with my calculated $R=\sigma_2^C/\sigma_2^L$, I converted their dipole length results to σ_2^C. My results are higher than Pindzola and Kelly's[8] by as much as a factor of 2 at shorter wavelengths. While it is well known that the single-photon ionization cross section of Ar $(3p)^6$ calculated in a one-electron model differs by a factor of 2 when compared with accurate many-body-theory calculations such as those of Pindzola and Kelly,[8] this is unlikely to be relevant to the factor of 2 here, as near threshold (1375 Å) the calculations are in good agreement. Pindzola and Kelly[8] did their calculation in LS coupling. In their calculation the $4s'$ resonance is shifted over to the $4s$ resonance and the $3d'$ resonance is shifted to the $3d$ resonance. To check the effect of the difference in coupling schemes, I readjusted E_0 in each Green's function to relocate the $4s'$ resonance at the $4s$, and $3d'$ and $3d''$ at the $3d$, i.e., an LS coupling calculation. The results are shown as open circles in Fig. 2. The choice of coupling does not resolve the factor of 2 discrepancy. However, in adjusting E_0 in the Green's function so that the $4s$ and $4s'$ resonances are located at their experimental resonances in jl coupling, and at the $4s$ resonance in LS coupling, all the higher ns and ns' resonances are shifted to longer wavelengths. This shift could account for the discrepancy. These considerations suggest the two calculations are in reasonable agreement.

In Fig. 4, the Xe two-photon calculation, there is a step in both σ_2^C and R at 1843 Å. At longer wavelengths ionization to the $(5p)^5\ {}^2P_{1/2}$ channel is energetically forbidden. For $\lambda > 1843$ Å, Beutler structure[14] should be present with large and interesting resonance effects. They have been neglected in these calculations.

IV. THREE-PHOTON IONIZATION CROSS SECTIONS

The cross section for three-photon ionization and the ratio $R=\sigma_3^C/\sigma_3^L$ for Ne, Ar, Kr, and Xe are shown in Figs. 5–8, respectively. Unlike the two-photon case, $R=\sigma_3^C/\sigma_3^L$ shows considerable variation. Table V lists expressions for R at the eight resonances. When matrix elements to continuum states with $l_f=4$ dominate those with $l_f=0$ and 2, $R=2.5$. At resonances with $J_2=0$ (p'' and p''''), $R=0$. At f and f'' resonances there can be structure in R due to final-state ($l_f=2, 4$) channel competition. At f', p, p', and p''' resonances, structure in R can arise from both final-state channel competition and interference of matrix elements.

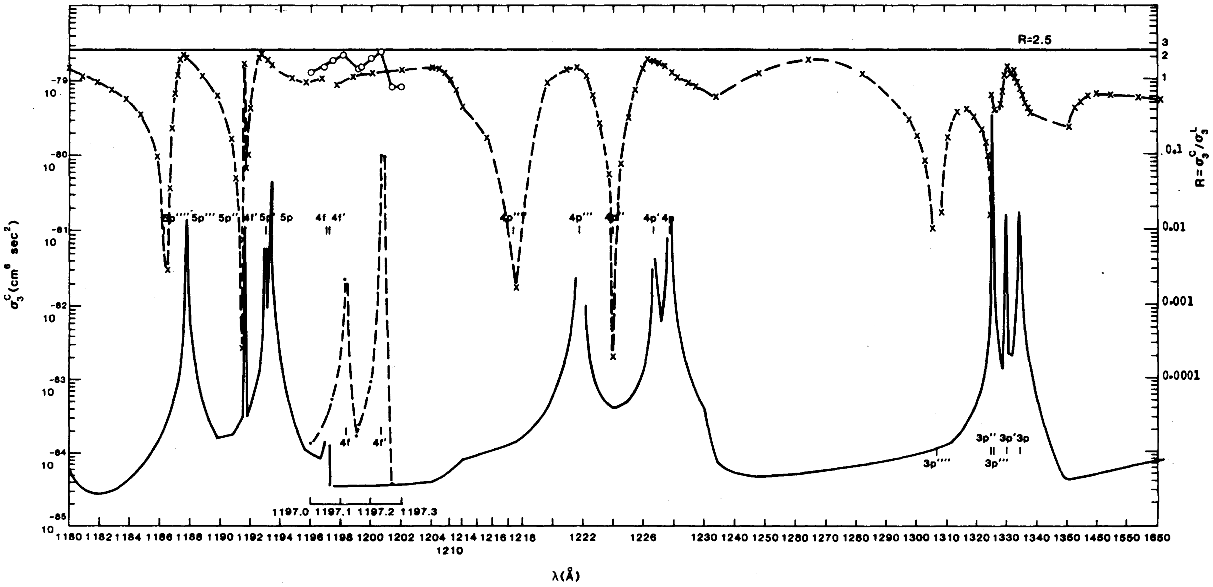

FIG. 5. The cross section for three-photon ionization with circularly polarized light and $R=\sigma_3^C/\sigma_3^L$ for Ne. The wavelength scale has been expanded and contracted to show resonance structure. The dashed curve for σ_3^C and open circles connected by solid lines for R show the $4f$ and $4f'$ resonances on a scale of 0.02 Å/division.

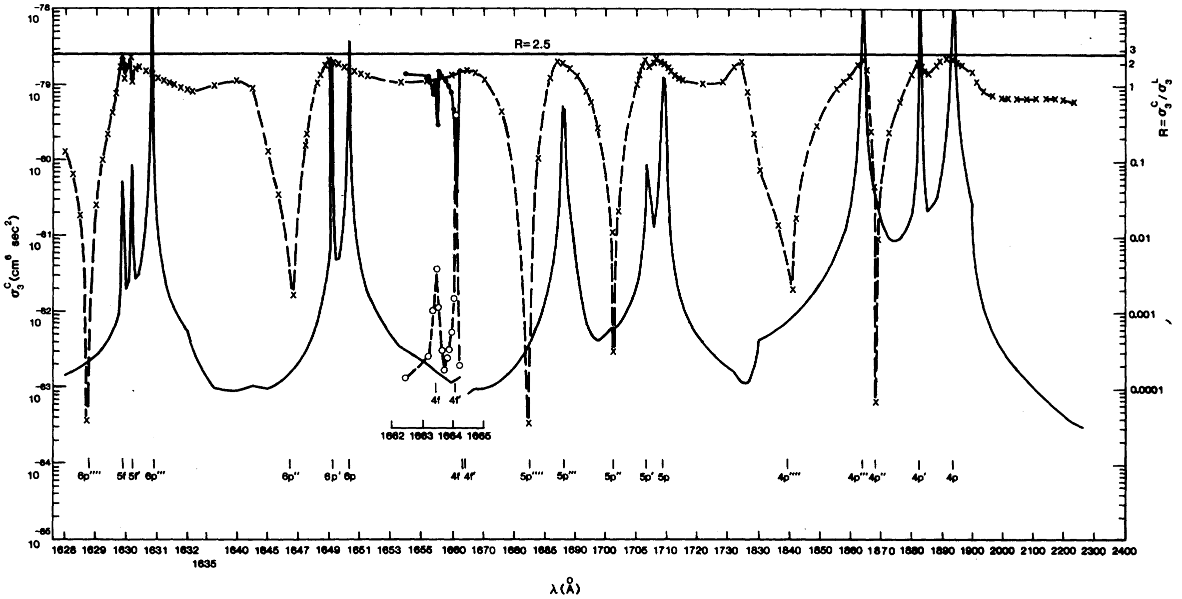

FIG. 6. Same as Fig. 5 for Ar. The $4f$–$4f'$ resonance structure is inseted on a scale of 0.2 Å/division with $\sigma_3^C(R)$ shown as open (closed) circles connected by dashed (solid) lines.

The three-photon cross section for circularly polarized light in Ne is shown in Fig. 5. The cross section near the $4f$–$4f'$ resonance is shown as a dashed curve on an expanded scale (0.02 Å/division) and the expanded scale R values are shown as open circles connected by solid lines. In Fig. 6 for Ar, the cross section for circularly polarized light and R values near the $4f$–$4f'$ resonances is seen clearly on a scale of 0.2 Å/division. An interesting feature for three-photon ionization in all the noble gases is the extent to which the R value differs from its maximum 2.5. In two-photon ionization, except for Ne, away from a resonance the ratio was $R \approx 1.5$.

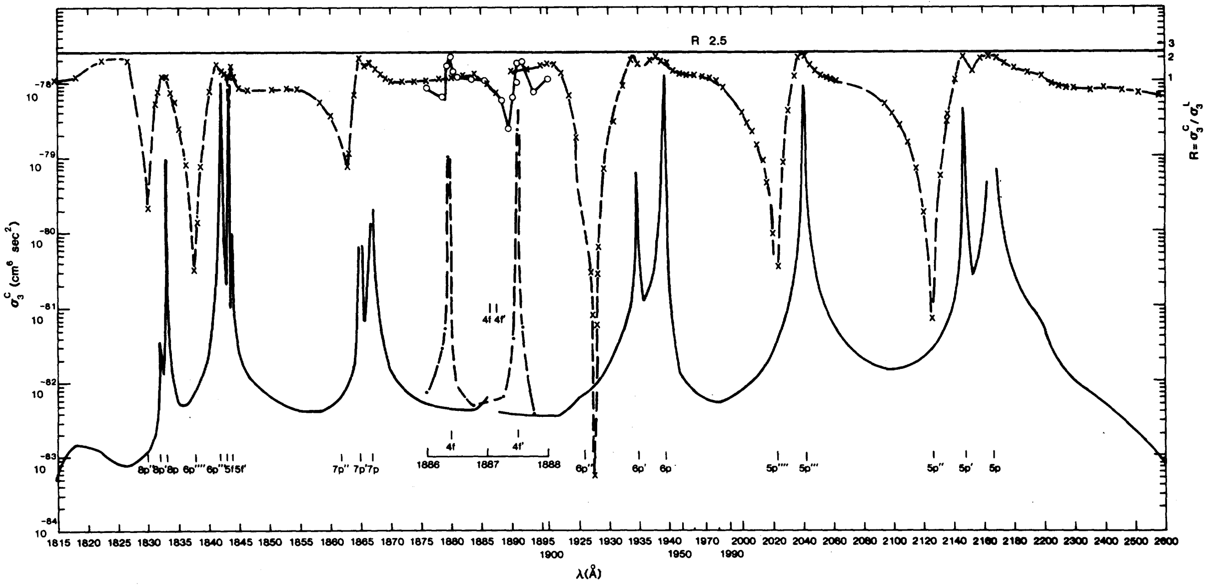

FIG. 7. Same as Fig. 5 for Kr. The $4f$–$4f'$ resonance structure is inseted on a scale of 0.1 Å/division with σ_3^C shown as a dashed curve and R as open circles connected by solid lines.

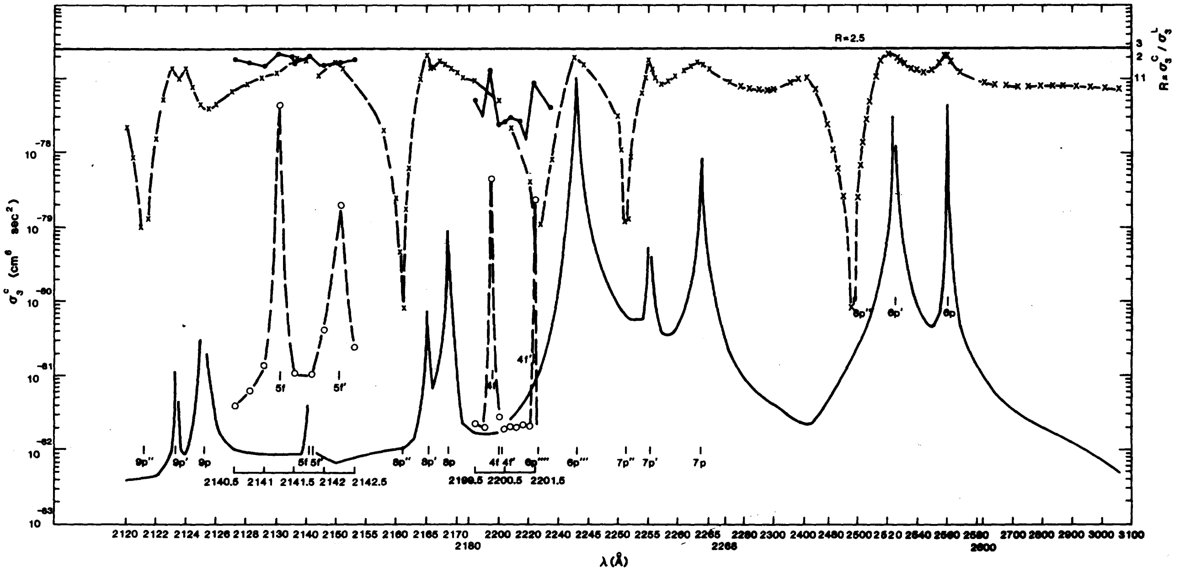

FIG. 8. Same as Fig. 5. The $4f$-$4f'$ and $5f$-$5f'$ resonance structures are inseted on scales of 0.2 and 0.1 Å/division; $\sigma_3^C(R)$ is shown as open (solid) circles connected by dashed (solid) lines.

V. DISCUSSION

The two-photon ionization calculations in Ar differed from those of Pindzola and Kelly[8] by as much as a factor of 2. An immediate question is the reliability of these calculations for Kr and Xe. This can only be assessed via other calculations and experiments. However, for Xe, I earlier used the

TABLE V. Values for $R=\sigma_3^C/\sigma_3^L$ at eight resonances in terms of matrix elements.

Designation	$J_f, l_2K_2J_2$	Ratio
f	$\frac{3}{2},3\frac{5}{2}\,2$	$\dfrac{\frac{5}{2}[Q\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,4)^2+\frac{29}{1350}Q(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)^2]}{[Q(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,4)^2+\frac{241}{270}Q(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)^2]}$
f'.	$\frac{3}{2},3\frac{3}{2}\,2$	$\dfrac{\frac{5}{2}[\vert Q(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,4)+Q(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,4)\vert^2+\frac{3}{200}\vert Q(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,2)+\frac{1}{5}Q(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2)\vert^2]}{[\vert Q(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,4)+Q(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,4)\vert^2+(\frac{37}{40}\vert Q(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,2)+\frac{1}{5}Q(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2)\vert^2]}$
f''	$\frac{1}{2},3\frac{5}{2}\,2$	$\dfrac{\frac{5}{2}[Q(\frac{1}{2},\frac{3}{2},\frac{5}{2}\,2,4)^2+\frac{1}{300}Q(\frac{1}{2},\frac{3}{2},\frac{5}{2}\,2,2)^2]}{[Q(\frac{1}{2},\frac{3}{2},\frac{5}{2}\,2,4)^2+\frac{59}{60}Q(\frac{1}{2},\frac{3}{2},\frac{5}{2}\,2,2)^2]}$
p	$\frac{3}{2},1\frac{5}{2}\,2$	$\dfrac{\frac{29}{12}[P(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)+\frac{1}{5}S(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)]^2}{[\vert P(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)+\frac{1}{5}S(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,2)^2+\frac{5}{6}\vert P(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,0)+\frac{1}{5}S(\frac{3}{2},\frac{3}{2},\frac{5}{2}\,2,0)\vert^2]}$
p'	$\frac{3}{2},1\frac{3}{2}\,2$	$\dfrac{\frac{189}{92}[\vert P(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2)+S(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,2)-\frac{4}{5}S(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2\vert^2}{[\vert P(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2)+S(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,2)-\frac{4}{5}S(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,2\vert^2+\frac{5}{46}\vert P(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,0)+S(\frac{3}{2},\frac{1}{2},\frac{3}{2}\,2,0)-\frac{4}{5}S(\frac{3}{2},\frac{3}{2},\frac{3}{2}\,2,0)\vert^2]}$
p''	$\frac{3}{2},1\frac{1}{2}\,0$	0
p'''	$\frac{1}{2},1\frac{3}{2}\,2$	$\dfrac{\frac{27}{11}\vert P(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2)+\frac{1}{5}S(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2)\vert^2}{[\vert P(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2)+\frac{1}{5}S(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2\vert^2+\frac{10}{11}\vert P(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2)+\frac{1}{5}S(\frac{1}{2},\frac{1}{2},\frac{3}{2}\,2,2)\vert^2]}$
p''''	$\frac{1}{2},1\frac{1}{2}\,0$	0

same central potential to calculate term splittings, Lande g factors, oscillator strengths, and lifetimes.[21] A comparison of these lifetimes calculations with other calculations and experiment may illuminate the reliability question. Table VI lists my calculated lifetimes for terms of the $(5p)^5\,(nl)$ configurations in Xe where $nl = 6s$, $6p$, and $5d$. For the $K, J = \frac{3}{2}$, 1 term of $(5p)^5(6s)$ the calculation is in excellent agreement with the measurement of Anderson[22] (fifth column), who determines lifetimes from linewidths; for the $K, J = \frac{1}{2}$, 1 the calculation and experiment differ by at least a factor of 2. This is a surprising result. For the $(5p)^5(6s)$ configuration jl and jj coupling lead to equivalent wave functions, and the calculations in Ref. 21 indicate almost perfect jj coupling. Condon and Shortley[23] show the transformation from LS to jj coupling as

$$\varphi(\tfrac{3}{2}, \tfrac{1}{2}, 1) = \sqrt{\tfrac{2}{3}}\;\varphi({}^1P_1) + \sqrt{\tfrac{1}{3}}\;\varphi({}^3P_1)\,,$$

$$\varphi(\tfrac{1}{2}, \tfrac{1}{2}, 1) = -\sqrt{\tfrac{1}{3}}\;\varphi({}^1P_1) + \sqrt{\tfrac{2}{3}}\;\varphi({}^3P_1)\,;$$

then the $\Delta S = 0$ selection rule leads to the two to one lifetime ratio.

For those terms of $(5p)^5(6p)$ for which measurements exist (columns 7 and 8 of Table VI) the tabulated values Allen *et al.*[24] are in excellent agreement with mine. These are essentially the values of Statz *et al.*[25] as corrected by Allen *et al.*[24] Reference 24 is essentially an experimental paper and their experimental results are listed in column 7. Two of their three measurements with small error bars are substantially larger than the calculated values. Jiminez *et al.*[26] have also measured

TABLE VI. Comparison of my calculated Xe excited-state lifetimes with other calculations and measurements.

Configuration	K	J	τ (ns) Calc	τ (ns) Ref. 22	τ (ns) Ref. 24	τ (ns) Ref. 24	τ (ns) Ref. 26	τ (ns) Ref. 27	τ (ns) Ref. 28
$(5p)^5(6s)$	$\frac{3}{2}$	1	3.5	3.8					
	$\frac{1}{2}$	1	7.4	3.2					
$(5p)^5(6p)$	$\frac{1}{2}$	1	38.0		47.0				
	$\frac{5}{2}$	2	35.7		45.0				
	$\frac{5}{2}$	3	29.9		33.0				
	$\frac{3}{2}$	1	32.3		35.0				
	$\frac{3}{2}$	2	27.6		27.1	33 ± 20			
	$\frac{1}{2}$	0	28.7		27.0	40 ± 12			
	$\frac{3}{2}$	1	28.0		28.7				
	$\frac{1}{2}$	1	23.1		24.8	43.5 ± 1.5	29.5 ± 3		
	$\frac{3}{2}$	2	26.4		28.0	29.0 ± 1.5	30.5 ± 3		
	$\frac{1}{2}$	0	21.9		23.0	38.5 ± 1.5	30.7 ± 1		
$(5p)^5(5d)$	$\frac{1}{2}$	0	670.0		2200.0				
	$\frac{1}{2}$	1	63.0		2700.0				
	$\frac{7}{2}$	4	1210.0		5250.0			1200.0	1330.0
	$\frac{3}{2}$	2	862.0		2500.0			2900.0	1020.0, 2100.0
	$\frac{7}{2}$	3	495.0		1365.0			1000.0	1170.0
	$\frac{5}{2}$	2	360.0		900.0			1700.0	1010 ± 50
	$\frac{5}{2}$	3	337.0		720.0				
	$\frac{3}{2}$	1	1.1		174.0				<350.0
	$\frac{5}{2}$	2	530.0						
	$\frac{3}{2}$	2	610.0						
	$\frac{5}{2}$	3	495.0						
	$\frac{3}{2}$	1	0.73						<350.0

the lifetimes of these three levels. For two there is excellent agreement with the calculations. For the $K, J = \frac{1}{2}, 0$, their measured value is 50% larger than the calculations.

For the lifetimes of the terms of the $(5p)^5(5d)$ configuration my calculations differed significantly from those tabulated in Ref. 24. For both sets of calculations agreement can be found with some of the measurements of Schlossberg and Javan[27] (column 9) and Davis and King[28] (column 10).

Given the sometime indirect nature and the difficulty of lifetime measurements, the differences between calculation and experiment shown in Table VI are not surprising. The agreement between the two sets of calculations for $(5p)^5(6p)$ is surprising in light of the disagreement for $(5p)^5(5d)$, as the calculations of Statz *et al.*,[24] as modified by Allen *et al.*,[23] are quantum defect method (QDM) calculations. While applying the QDM method to low-lying noble-gas excitations is dubious, it should be no worse for $(5p)^5(5d)$ than for $(5p)^5(6p)$.

VI. CONCLUSIONS

The two- and three-photon ionization cross sections in the noble gases, presented here, should prove a useful comparison with measurements made with short-wavelength lasers. In comparison with the many-body–theory calculations of Pindzola and Kelly on two-photon ionization in Ar, these Green's-function calculations differed by as much as a factor of 2. However, it is likely this difference is an artifact of the computational approximation, and, after accounting for the artifacts, it appears the two calculations are in reasonable agreement.

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[1] J. S. Bakos, Adv. Electron. Electron Phys. 36, 57 (1974).

[2] P. Lambropoulos, in *Advances in Atomic and Molecular Physics*, edited by D. R. Bates and B. Bederson (Academic, New York, 1976), Vol. 12.

[3] N. R. Isenor, in *Multiphoton Processes*, edited by J. H. Eberly and P. Lambropoulos (Wiley, New York, 1978).

[4] J. A. Armstrong, P. Esherick, and J. J. Wynne, Phys. Rev. A 15, 180 (1977).

[5] P. Esherick, Phys. Rev. A 15, 1920 (1977).

[6] Early work is reviewed in C. Grey Morgan, Rep. Prog. Phys. 38, 621 (1975). For later results see L. A. Lompre, G. Mainfrey, G. Manus, S. Repoux, and J. Thebault, Phys. Rev. Lett. 36, 949 (1976); and L. A. Lompre, G. Mainfrey, C. Manus, and J. Thebault, Phys. Rev. A 15, 1604 (1977).

[7] G. A. Victor, Proc. Phys. Soc. London 91, 825 (1967).

[8] M. S. Pindzola and H. P. Kelly, Phys. Rev. A 11, 1543 (1975).

[9] B. Ritchie, Phys. Rev. A 16, 2080 (1977).

[10] K. Aron and P. M. Johnson, J. Chem. Phys. 67, 5099 (1977).

[11] F. H. M. Faisal, R. Wallenstein, and H. Zacharias, Phys. Rev. Lett. 39, 1138 (1977).

[12] C. K. Rhodes and P. W. Hoff, in *Eximer Lasers*, edited by C. K. Rhodes (Springer, New York, 1979).

[13] J. Rice, 7th Winter Colloquium in High Power Visible Lasers, Park City, Utah (1977) (unpublished).

[14] H. Beutler, Z. Phys. 93, 177 (1935).

[15] H. B. Bebb, Phys. Rev. 149, 25 (1966).

[16] M. R. Teague, P. Lambropoulos, D. Goodmanson, and D. W. Norcross, Phys. Rev. A 14, 1057 (1976).

[17] E. J. McGuire, Phys. Rev. A 23, 186 (1981).

[18] C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, D. C.), Vol. I (1949), Vol. II (1952), Vol. III (1958).

[19] The dependence of the numerical factor on the choice of energy units is discussed in Ref. 17.

[20] Detailed calculation of the expressions for two- and three-photon ionization are found in Sandia Research Report, SAND80-1974 (unpublished).

[21] E. J. McGuire, Sandia Research Report, SAND76-0196 (unpublished).

[22] D. K. Anderson, Phys. Rev. 137, A21 (1965).

[23] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1963).

[24] L. Allen, D. G. C. Jones, and D. G. Schofield, J. Opt. Soc. Am. 59, 842 (1969).

[25] H. Statz, C. L. Tang, and G. F. Koster, J. Appl. Phys. 34, 2625 (1963).

[26] E. Jiminez, J. Campos, and C. Sanchez del Rio, J. Opt. Soc. Am. 64, 1009 (1974).

[27] H. R. Schlossberg and A. Javan, Phys. Rev. Lett. 17, 1242 (1966); Phys. Rev. 150, 267 (1966).

[28] C. C. Davis and T. A. King, J. Quant. Spectrosc. Radiat. Transfer 13, 825 (1973).