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# Observation of Electric Quadrupole Transitions in Multiphoton Ionization*

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Two flashlamp-pumped, tunable, dye lasers have been utilized to study three-photon ionization in atomic sodium. The resulting ion yield shows large peaks at laser frequencies which correspond to electric quadrupole transitions. This is the first direct observation of electric quadrupole effects in multiphoton ionization. The $3p\ ^2P_{3/2} \rightarrow 4f$ matrix element is determined, and the fine-structure splitting of the $5p$ state is measured.

Previous work on tunable-laser excitation of atoms, with one exception,[1] has dealt exclusively with electric dipole processes. Various experiments have looked at both single-photon and two-photon resonances, and have monitored fluorescence,[2-6] stimulated emission,[7,8] or ionization.[9-13] Experiments on multiphoton ionization with high-intensity fixed-frequency lasers[14-16] have heretofore included only electric dipole transitions in the models for interpretation.[17] Recently, however, calculations of Lambropoulos, Doolan, and Rountree[18] on two-photon ionization in lithium have indicated that electric quadrupole transitions are dominant over the nonresonant dipole background for certain frequencies. Using two tunable dye lasers, we have measured the three-photon ionization rate of sodium as a function of laser frequencies, and have observed large resonances due to such transitions. Our results emphasize the necessity of including weak dipole-forbidden transitions in the interpretation of experiments using strong narrow-band sources.

For the results shown here, one laser is tuned to one of the $3s \rightarrow 3p$ transitions, while the second laser frequency is scanned to map out a higher transition, such as $3p \rightarrow 4f$, and ionization from this upper state is measured. The partial energy-level diagram in Fig. 1 shows the transitions which we excite. Resonances due to the $3p \rightarrow 5p$ and $3p \rightarrow 4f$ electric quadrupole transitions rise more than 2 orders of magnitude above the instrumental background (see Fig. 2) and an estimated 6 to 8 orders of magnitude above the nonresonant dipole background, depending upon the line in question. Careful measurements of the $3p \rightarrow 4d$ resonance provide a well-understood electric dipole comparison process.

In sodium, since the $3s \rightarrow 3p$ transitions have large oscillator strengths, it is necessary to pump them very weakly to avoid line-shape problems due to stimulated emission.[19,20] The quadrupole resonance, in contrast, may be excited with a much more intense laser before the onset of saturation. In this case, the ion yield is given

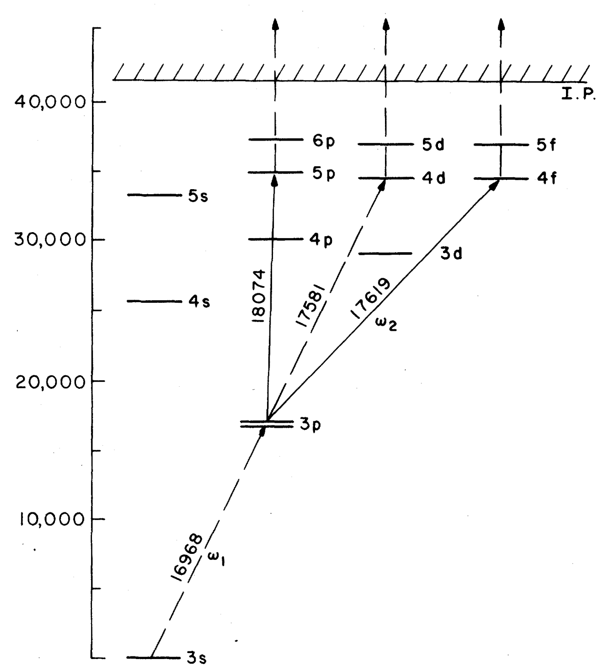


FIG. 1. Partial energy-level diagram of sodium, showing dipole transitions by broken lines and quadrupole transitions by solid lines. The vertical scale is energy in units of inverse centimeters.

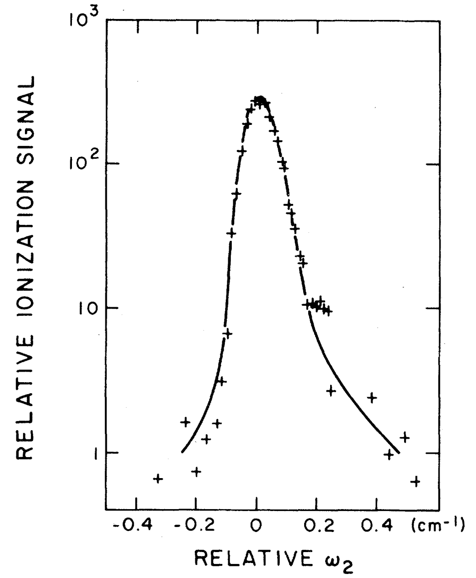


FIG. 2. The $3s \rightarrow 3p\,^2P_{3/2} \rightarrow 4f \rightarrow e^-$ resonance is shown as a function of $\omega_2$.

by

$$W \propto I_1 I_2^2 \left| \sum_{n,m,\vec{k}} \frac{\langle \epsilon \vec{k} | V_D | n \rangle \langle n | V_Q | m \rangle \langle m | V_D | i \rangle}{(\omega_1 + \omega_2 - \omega_n + i\Gamma_n)(\omega_1 - \omega_m + i\Gamma_m)} \right|^2 ,$$

where $I_1$ and $I_2$ are the experimental laser intensities and $\omega_1$ and $\omega_2$ the frequencies of the lasers which excite states $m$ and $n$, respectively. $V_D$ and $V_Q$ are the electric-dipole and electric-quadrupole operators; $|i\rangle$, $|m\rangle$, and $|n\rangle$ are the ground state and first and second excited states of the atom, respectively; the atomic frequencies are $\omega_n = \hbar^{-1}(E_n - E_i)$; and $\Gamma_n$ are the widths. This equation, which is in a form specifically applicable to this experiment, may also be written[20] in a form which isolates the atomic effects in a single quantity called the generalized three-photon cross section $\hat{\sigma}_3$; then $W \propto I_1 I_2^2 \hat{\sigma}_3$. In addition to containing the atomic matrix elements and resonance frequencies, however, $\hat{\sigma}_3$ also depends upon the laser intensities through the level widths $\Gamma_n$, because when the incident intensity is high enough, the lifetime of an atom in a particular state may be determined by stimulated emission or subsequent absorption, rather than simply spontaneous emission. Then the relevant (intensity-dependent) width must be included in the denominator if one is to infer ratios of atomic matrix elements from observed peak heights in the ion-yield spectrum.

The experiment is performed in a crossed-beam geometry using a collimated sodium beam and two flashlamp-pumped dye lasers. The 5-mm-diam sodium beam, whose divergence is 10 mrad, is intersected at right angles by the combined and focused beams of the two lasers. The laser-spot diameter is approximately 250 $\mu$m, and the sodium-beam density about $10^{10}/\text{cm}^3$. Ions are swept from the parallel-plate interaction region by a field of about 40 V/cm and detected by a particle multiplier. Time-of-flight discrimination allows separation of monatomic ions from the small diatomic-ion background. Laser powers are monitored by photodiodes which are periodically calibrated against a thermopile to allow absolute laser pulse energies to be extracted. The neutral sodium beam is monitored by a Langmuir-Taylor detector. An on-line computer logs all parameters on a shot-by-shot basis and normalizes the ion yield at each point. The lasers, whose repetition rate is 2.5 pulses/sec, have pulse durations of 700 nsec (full width at half-maximum) and peak powers up to 40 kW; they are synchronized to within the jitter of the flash lamps (typically 100 nsec) and their time overlap is monitored. Data are discarded from those shots for which the pulse overlap is unsuitable. Each laser is tuned with a diffraction grating and one or more etalons, allowing linewidths down to 1 GHz (0.03 $\text{cm}^{-1}$). In these experiments, laser 1 had a linewidth of 2 $\text{cm}^{-1}$ and was highly attenuated to ensure a low intensity on the atomic line, while laser 2 had a lindwidth of 0.03 $\text{cm}^{-1}$ in order to probe finely the $3p \rightarrow 5p$ and $3p \rightarrow 4f$ transitions. Both lasers were linearly polarized.

We easily resolve the fine structure of the $5p$ state in excitation from the $3p\,^2P_{3/2}$, and the absence of the $3p\,^2P_{1/2} \rightarrow 5p\,^2P_{1/2}$ peak provides confirmation of the electric quadrupole nature of the transition ($\Delta J = 0$ forbidden for $\frac{1}{2} \rightarrow \frac{1}{2}$).[21,22] Figure 3 shows the experimental result for the fine-structure splitting, $2.45 \pm 0.06$ $\text{cm}^{-1}$, where the error is due to the limit of resettability of the etalon angle, and to the determination of its free spectral range. This measurement compares to tabulated values which range from 2.47[23] to 2.52 $\text{cm}^{-1}$.[24] As a further test of the electric quadrupole nature of the transitions, the ion yield for the process $3s \rightarrow 3p\,^2P_{3/2} \rightarrow 4f \rightarrow e^-$ was measured as a function of external electric field. Variation of the interaction-region sweep field over a factor of 3 produced no change in the ion yield, whereas a field-induced process such as Stark mixing of the $4d$ with the $4f$ should have exhibited an $E^2$ dependence.

The measured ratio of the ion yields for ioniza-

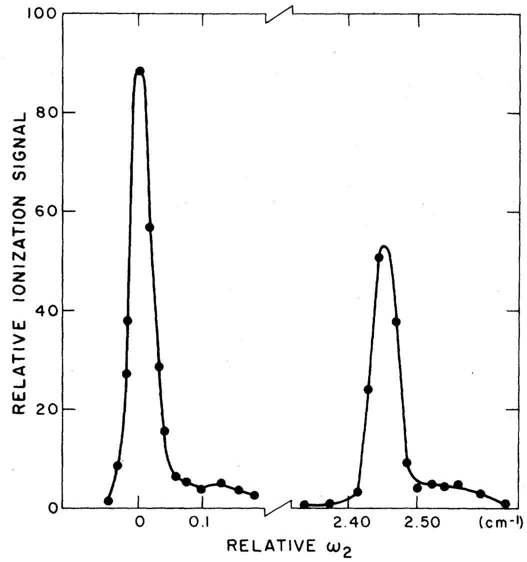


FIG. 3. Three-photon-ionization generalized cross section is plotted against frequency of the second laser. Exciting the $3p\ ^2P_{3/2}$ level by one laser whose frequency is fixed, the second laser scans frequency from the $3p\ ^2P_{3/2}$ to the $5p\ ^2P_{3/2,\,1/2}$ levels.

tion via the $4d$ or $4f$ channel[25] allows comparison of our results to the calculation of Tull *et al.*[26] for the quadrupole transition strength. With use of the known dipole transition strength, bound-free matrix elements from quantum-defect theory,[27] and the fact that the level widths differ negligibly (< 10%) under our experimental conditions, a theoretical comparison value of $4.5\times10^{-6}$ is obtained.[28] The experimental values were taken under similar conditions to the data in Fig. 2, and except for the $10^4$ attenuation of laser 2, $4d$ and $4f$ were measured with constant parameters —$I(\omega_1)$, sodium flux, focal volume, and laser linewidth. Since the experimental ratio of $6\times10^{-6}$ has a factor of 2 uncertainty, we are in good agreement with the calculated matrix element: $|\langle 3p|r^2|4f\rangle| = 1.05\times10^{-15}$ cm$^2$.

More detailed data on these processes will provide experimental determination of bound-free matrix elements or their ratios.[27,29] For example, relative measurements of the ratio of the generalized cross sections $\hat{\sigma}_3$ for the three $3s \rightarrow 3p \rightarrow 5p \rightarrow k$ peaks will yield a measurement of the ratio of the $s$ to $d$ outgoing-wave matrix elements. In addition, measurement of the cross sections with light of different polarizations provides useful information: The ratio of $\hat{\sigma}_3(3s \rightarrow 3p \rightarrow 4f \rightarrow k$; linear light polarization) to $\hat{\sigma}_3(3s \rightarrow 3p \rightarrow 4f \rightarrow k$; circular light polarization) will allow determination of the hitherto unmeasured outgoing $g$-wave matrix element relative to the $d$-wave matrix element. Whether the measurement is of fluorescence or ionization, this technique will allow determination of electric-quadrupole transition strengths, providing a sensitive and previously unavailable test of calculated matrix elements. It will render more accessible a study of the fine and hyperfine structure of bound states of high orbital angular momentum, and allow measurement of ionization matrix elements of such states into high-$l$ partial waves. In addition to their angular momentum properties, the very weakness of electric-quadrupole transitions makes them effective narrow probes for selective excitation or studies of perturbed atomic states. For example, we are presently using ionization via the $4f$ state to study the splitting of the $3p\ ^2P_{3/2}$ state of sodium in laser fields more intense than those used in the present work.

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# Low-Frequency Stability of Astron Configurations*

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A sufficient condition is obtained for stability of low-frequency kink perturbations of an astron-type particle ring embedded in a dense low-temperature plasma.

There is renewed interest in the use of intense beams of high-velocity particles in toroidal astron configurations.[1-3] Such beams may allow the confinement and heating of a plasma at fusion temperatures and densities. It is desired to have the directed kinetic energy density of the particle beam as large a fraction ($\beta$) as possible of the energy density of the external magnetic field. Stability requirements on the low-frequency magnetohydrodynamic motion of the beams are expected to impose upper limits on the allowed $\beta$ values.

In the astron system first proposed,[1] the external magnetic mirror field is canceled on axis by the poloidal self-magnetic-field of a beam of relativistic electrons.[4] The use of a beam of high-velocity nonrelativistic ions[2,3] has the important advantage that the incoherent synchrotron radiation is negligible. Field cancelation on axis provides a closed magnetic field suitable for confining a high-pressure plasma, and it requires that the kinetic energy density of the particle beam be a fraction $\beta > 1$ of the energy density of the external magnetic mirror field. The magnetohydrodynamic stability of an astron configuration has been previously studied[5] with, however, the restrictive assumption that the relativistic electrons may be treated as rigid and immobile. A detailed analysis of the low-frequency stability of astron- and tokamak-type particle rings including the beam dynamics has recently been completed,[6] and I summarize here some of the results for astron configurations.

The analysis is done in linear cylindrical geometry, with coordinates $(r, \theta, z)$, where the $z$ axis coincides with the axis of the unperturbed beam. This assumes that the inverse aspect ratio is small, $a/R \ll 1$, where $a$ is the minor radius of the torus and $R$ is the major radius. The toroidal nature of the actual system enters through the requirement that the perturbation quantities be periodic over the axial length $2\pi R$. Hence the perturbation quantities have a $\theta, z$ dependence of the form $\exp(im\theta + ikz)$, with the wave number $k = n/R$, where $n$ and $m$ are integers. Approximations made in describing the beam dynamics restrict consideration to $m = \pm 1$, $n \neq 0$. Toroidal effects on the beam-particle dynamics are discussed in the final paragraph.

A dense plasma is assumed to fill the region occupied by the particle beam. The plasma may