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MULTIPHOTON IONIZATION OF CESIUM ATOMS ABOVE AND BELOW THE TWO-PHOTON IONIZATION THRESHOLD*

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ABSTRACT

Two- and three-photon ionization processes in cesium atoms using a single pulsed-dye laser and a cesium atomic beam have been investigated. Photoelectron angular distributions have been measured for two-photon resonant, three-photon ionization via the nd states (n = 12, 15, and 21), for two-photon nonresonant transitions over the photoelectron energy range from ~26-100 meV, and for "quasi-free-free" transitions.

INTRODUCTION

In the last several years research in the field of multiphoton ionization (MPI) has rapidly advanced. A number of review articles have been published concerning theory and experiment for both resonant and nonresonant MPI processes (see Refs. 1-4 and references cited therein). In particular, the study of photoelectron angular distributions utilizing MPI processes in atoms has received considerable attention.[5-8] Photoelectron angular distributions depend on the nature of the quantum states involved and hence prove important in obtaining information about both the initial bound state and final continuum states of the photoejected electron and the microscopic parameters of the particular system under study. To our knowledge, photoelectron angular distribution measurements via MPI of cesium in an atomic beam are limited to MPI via 7^2P states.[9]

In this paper we report measurements of photoelectron angular distributions from cesium atoms for (1) two-photon resonant three-photon ionization via the n = 12, 15, and 21d states, (2) two-photon nonresonant ionization, and (3) "quasi-free-free" transitions, using a single laser and a cesium atomic beam.

EXPERIMENTAL

The experimental apparatus employed in the present study is shown schematically in Fig. 1. It consists of a frequency tunable dye laser, the polarization optics, and the vacuum chamber. The latter includes the atomic beam assembly and the electron energy analyzer and detector. The dye laser was pumped by a Nd:YAG laser

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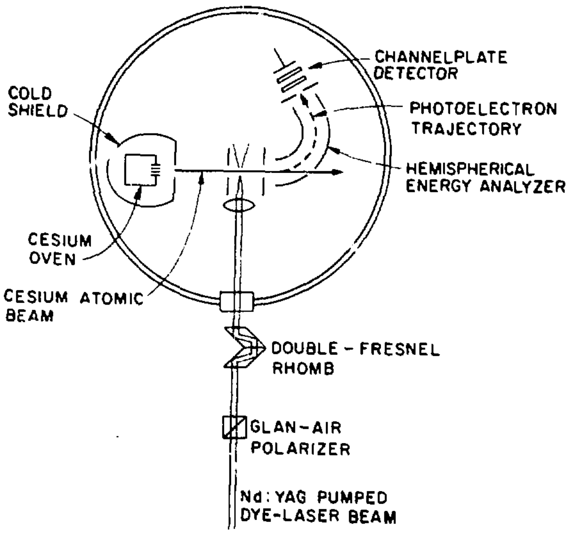

Fig. 1. Experimental apparatus.

(Quanta Ray) and had a pulse duration of 8 ns and a bandwidth of ~0.02 nm in the spectral region of interest ($610 \leq \lambda \leq 685$ nm). The laser beam was linearly polarized using a Glan-air polarizer and was focused by a 35 mm focal length lens into the cesium atomic beam ~1 cm from the entrance aperture of the energy analyzer. The laser beam crossed the cesium atomic beam at 90° and the interaction volume defined solely by the laser beam focus was estimated to be $\sim 10^{-4}$ cm^3. The laser power density at the focal point was $\sim 10^8$ W/cm^2. The polarization direction of the laser light was rotated using a double-Fresnel rhomb.

Metallic cesium was evaporated in a resistively heated stainless steel oven. The cesium atoms exited the oven through a multichannel array and were directed into the entrance of the energy analyzer after passing through a water-cooled baffle and a second 3-mm-dia defining aperture. The cesium beam finally passed through a small hole in the back of the analyzer. The cesium atom number density in the interaction volume was estimated to be $\sim 10^{13}$ atoms/cm^3 at an oven temperature of 120°C.

The electrons produced by MPI of the cesium and ejected perpendicular to the propagation vector of the laser beam were energy analyzed by a spherical sector electrostatic energy analyzer and were detected by a dual channelplate charged particle detector. The amplified output of the detector was sampled by a boxcar integrator (Princeton Applied Research, model 162) and plotted on a x-y recorder as a function of the laser excitation wavelength or the angle θ between the polarization direction of the laser light and the direction of the detected photoelectrons. The analyzer had an acceptance angle of ~3° and a resolution of ~0.4 eV at a transmission energy of ~38 eV. This resolution was sufficient for the present work where either single or well separated (~2 eV) atomic peaks were being studied. Space charge broadening was minimized by reducing both the laser power density ($\sim 10^8$ W/cm^2) and the cesium atom number density ($\sim 10^{13}$ atoms/cm^3).

Normal operation was under electric field-free conditions and the earth's magnetic field was compensated for by using 40-cm radius Helmholtz coils. An electrode located ~1 cm from the interaction volume and in front of the energy analyzer allowed the application of an external electric field.

Photoelectron angular distributions were obtained by monitoring the photoelectron intensity as a function of angle Θ. Spectra of photoelectron signal intensity as a function of the laser excitation wavelength at a fixed transmission energy of the energy analyzer were also measured.

RESULTS AND DISCUSSION

Referring to the MPI theory of photoelectron angular distributions, we note that the differential cross section for photoelectron emission can be represented by

$$\frac{d\sigma^{(N)}(\lambda,\Theta)}{d\Omega} = \frac{\sigma^{(N)}_{TOT}(\lambda)}{4\pi} \sum_{i=0}^{N} \beta_{2i} P_{2i} (\cos \Theta) \tag{1}$$

where N is the number of absorbed photons, $\sigma^{(N)}_{TOT}$ is the generalized total cross section for ionization at wavelength λ, P_{2i} (cos Θ) are the Legendre polynominals of the order 2i, and β_{2i} are the asymmetry parameters. The β_{2i} are functions of the microscopic atomic properties of the system under study and in general depend on the laser light intensity. However for the power densities (~10^8 W/cm^2) used in the present experiment, the β_{2i} parameters were independent of the laser intensity. For mathematical convenience and analysis of the experimental data, Eq. (1) can be written as a summation of even powers of cos Θ and expressed as

$$\frac{d\sigma^{(N)}(\lambda,\Theta)}{d\Omega} = \frac{\sigma^{(N)}_{TOT}(\lambda)}{4\pi} A_N \sum_{i=0}^{N} \beta_{2i} \cos^{2i}(\Theta) \tag{2}$$

where A_N is a normalizing constant corresponding to N photon ionization.

Two-Photon Resonant Three-Photon Ionization via the nd States

Figure 2(a) shows the two-photon resonant three-photon ionization scheme used to study the nd states of cesium. Using this excitation scheme, photoelectron angular distributions were measured for the n = 12d, 15d, and 21d states and the result for n = 12d is shown in Fig. 3. The solid line through the experimental points was obtained by a least squares fitting procedure performed to Eq. (2). The minimum value of χ^2 was used as a criterion for the best fit. This gave an intensity distribution containing powers of cos Θ up to the sixth order and the β coefficients calculated using this method were $\beta_0 = 1$, $\beta_2 = 15.09$, $\beta_4 = -39.57$, and $\beta_6 = 38.77$.

Nonresonant Two-Photon Ionization

The excitation scheme used for two-photon nonresonant ionization of cesium atoms is shown in Fig. 2(b). Figure 4 shows the photoelectron signal as a function of laser excitation wavelength ($630 \leq \lambda \leq 646$ nm) for: E = 0 (spectrum I), E = 0.1 V/cm

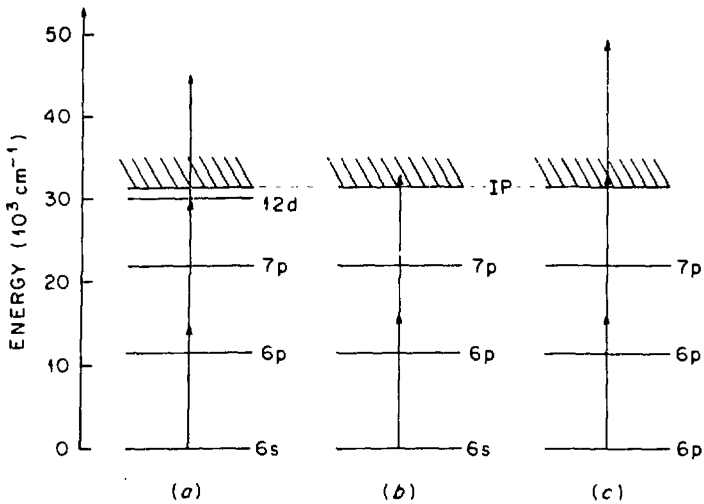

Fig. 2. Energy level diagrams for multiphoton ionization of cesium atoms (a) two-photon resonant three-photon ionization via the nd state, (b) two-photon nonresonant ionization, and (c) quasi-free-free transitions.

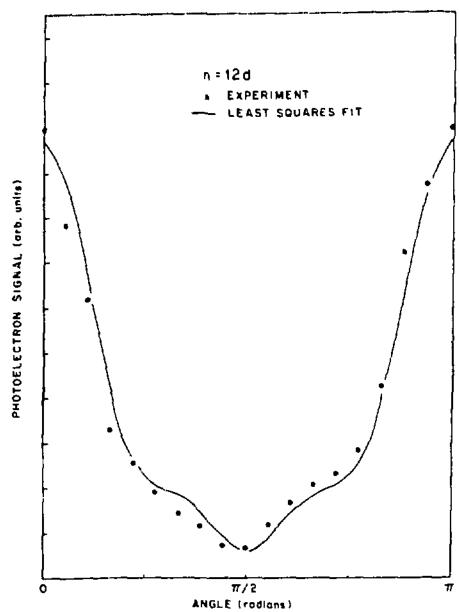

Fig. 3. Photoelectron angular distributions for two-photon nonresonant three-photon ionization via the n = 12d state in cesium atoms.

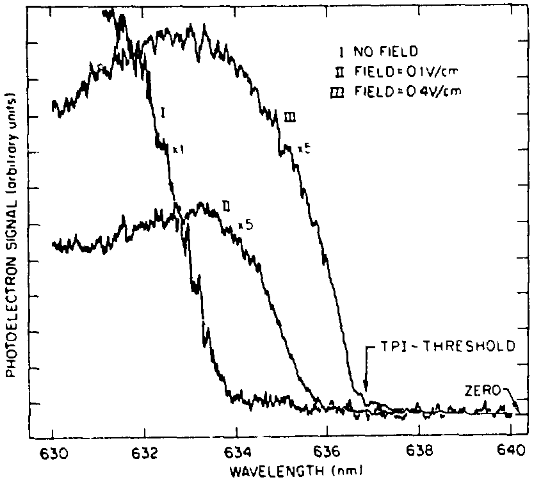

Fig. 4. Spectrum of photoelectron signal as a function of laser excitation wavelength for two-photon nonresonant ionization in cesium atoms. The arrow indicates the position of the two-photon ionization (TPI) threshold which corresponds to one-photon energy of 1.946 eV.

(spectrum II), and E = 0.4 V/cm (spectrum III) where the electric field was used to repel the photoelectrons with the energy analyzer. In spectrum I of Fig. 4 it was observed that the ionization threshold was blue shifted by ~140 cm^{-1}, but the application of a small dc field (spectra II and III) clearly moved this threshold towards the two-photon ionization (TPI) threshold which corresponds to a one-photon energy of 1.940 eV. This leads to the conclusion that this apparent shift in the photoelectron threshold can be attributed to the inability of our detection system to detect electrons with energy less than ~20 meV. Note that the 0.4 eV resolution of the energy analyzer prevents the simultaneous transmission of photoelectrons from both two- and three-photon ionization processes since the difference in photoelectron energies is ~2 eV. This explains the absence of the three-photon ionization signal below the two-photon ionization threshold (λ = 636.8) in the spectra of Fig. 4.

Photoelectron angular distributions were measured at seven excitation wavelengths, λ = 632.5, 630.5, 628.5, 626.5, 624.5, 622.5, 620.0 nm corresponding to a photoelectron energy range from ~26-100 meV. A typical distribution is shown in Fig. 5 where λ = 622.5 nm and the photoelectron energy is ~89 meV. The solid line through the experimental data points is a least squares fit to Eq. (2). This gives an intensity distribution containing $\cos^2\theta$ and $\cos^4\theta$ terms and the β parameters calculated using this method are $\beta_0 = 1$, $\beta_2 = -3.72$ and $\beta_4 = 5.47$. It was observed that if all the photoelectron angular distributions were normalized to the same maximum value at $\theta = 0°$, then the peak at $\theta = \pi/2$ decreased as the exciting photon energy is increased. Preliminary theoretical analysis[10,11] of these measurements shows a good qualitative agreement, and a detailed quantitative comparison between theory and experiment will be published elsewhere.[12]

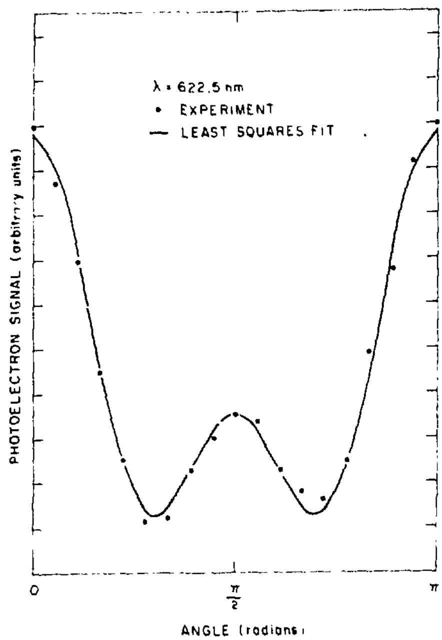

Fig. 5. Photoelectron angular distribution for two-photon nonresonant ionization in cesium atoms at a laser excitation wavelength λ = 622.5.

"Quasi-Free-Free" Transitions

The energy level scheme for the "quasi-free-free" transition studied in this experiment is shown in Fig. 2(c). Two photons (λ = 633.66 nm) were used to nonresonantly ionize a cesium atom and a third photon was absorbed in the continuum. The corresponding photoelectrons were observed at an energy of 3h −IP where h is laser photon (λ = 633.66 nm) energy and IP is the single photon ionization threshold (3.893 eV) of cesium. An angular distribution measured for this process is shown in Fig. 6 where the solid line through the experimental data is a least squares fit to Eq. (2). This procedure yields a sixth order polynominal of cos θ with the β coefficients given by $\beta_0 = 1$, $\beta_2 = 3.07$, $\beta_4 = -6.36$, and $\beta_6 = 8.47$. According to the selection rules in the electric dipole approximation, the final continuum state is composed of p and f partial waves, and the clear deviation from a $\cos^2\theta$ type function in the present photoelectron angular distribution indicates a substantial contribution from the $\cos^4\theta$ and $\cos^6\theta$ terms. Further experimental investigation is proceeding and efforts are being made to achieve a theoretical analysis for these measurements.

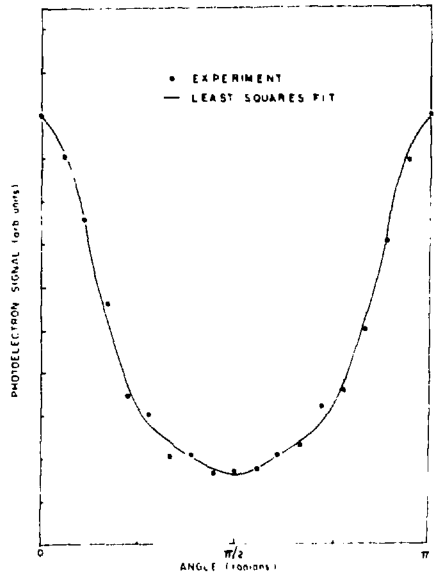

Fig. 6. Photoelectron angular distribution for two-photon nonresonant ionization with the absorption of an additional photon in the continuum (quasi-free-free transition) at a laser excitation wavelength λ = 633.66 nm.

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