Nonresonant Multiphoton Ionization of Alkaline Earth Atoms in Intense Laser Fields.

Dal-woo Kim
Louisiana State University and Agricultural & Mechanical College
INFORMATION TO USERS

The most advanced technology has been used to photograph and reproduce this manuscript from the microfilm master. UMI films the text directly from the original or copy submitted. Thus, some thesis and dissertation copies are in typewriter face, while others may be from any type of computer printer.

The quality of this reproduction is dependent upon the quality of the copy submitted. Broken or indistinct print, colored or poor quality illustrations and photographs, print bleedthrough, substandard margins, and improper alignment can adversely affect reproduction.

In the unlikely event that the author did not send UMI a complete manuscript and there are missing pages, these will be noted. Also, if unauthorized copyright material had to be removed, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, charts) are reproduced by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps. Each original is also photographed in one exposure and is included in reduced form at the back of the book. These are also available as one exposure on a standard 35mm slide or as a 17" x 23" black and white photographic print for an additional charge.

Photographs included in the original manuscript have been reproduced xerographically in this copy. Higher quality 6" x 9" black and white photographic prints are available for any photographs or illustrations appearing in this copy for an additional charge. Contact UMI directly to order.
Nonresonant multiphoton ionization of alkaline-earth atoms in intense laser fields

Kim, Dal-Woo, Ph.D.
The Louisiana State University and Agricultural and Mechanical Col., 1989

Copyright ©1990 by Kim, Dal-Woo. All rights reserved.
NONRESONANT MULTIPHOTON IONIZATION OF ALKALINE-EARTH ATOMS IN INTENSE LASER FIELDS

A Dissertation

Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Doctor of Philosophy

in

Department of Physics and Astronomy

by

Dal-Woo Kim

B.S., Seoul National University, 1973
M.S., Louisiana State University, 1987
August, 1989
ACKNOWLEDGEMENTS

I am grateful to Dr. L.F. DiMauro who undertook the burden of being my graduate advisor both on campus and outside of a college setting, at the Brookhaven National Laboratories in Long Island, N.Y. He has always been a model scientist and a perfect example for me as well as a guide and teacher for the last three years. It is a pleasure to express my gratitude to my group members M. Anselment, M. Saced, S. Fournier, and M. Courtney for their invaluable aid in doing my experiment. I am also indebted to the staff of Physics Department at LSU and Chemistry Department at BNL for their help and friendship. I would like to thank POSCO men for their help and encouragement during the course of this work. Finally, I wish to thank my family, Myo-Rae and Kwang-Youn, for their understanding and encouragement during my graduate work.
To Myo-Rae and Kwang-Youn
Table of Contents

Title Page .............................................................. 1
Acknowledgement Page .......................................... li
Dedication ............................................................. iii
Table of Contents ...................................................... iv
List of Tables .......................................................... vi
List of Figures .......................................................... vii
Abstract ............................................................... x

1. Introduction ......................................................... 1
2. Theoretical Considerations ....................................... 8
   2.1 Multiphoton Ionization Spectroscopy ....................... 8
   2.2 Ionization and Structure of Atoms in a Laser Field .... 10
   2.3 Angular Distribution of Photoelectrons .................. 24
   2.4 Energy Level Structure of Alkaline-Earth Atoms ....... 25
     2.4.1 Energy Levels of Calcium Atoms ..................... 27
     2.4.2 Energy Levels of Magnesium Atoms ................. 32
   2.5 Correlations of Two Excited Electrons ................. 35
3. Experimental Setup ............................................... 38
   3.1 The Laser and the Optical System ....................... 38
   3.2 Vacuum System ............................................. 43
   3.3 Time of Flight Spectrometer ............................ 46
   3.4 Electron and Ion Detection System ..................... 49
3.5 Data Acquisition and Analysis ........................................... 51

4. Nonresonant Multiphoton Ionization .................................. 55
   of Alkaline-Earth Atoms
   4.1 Calcium ........................................................................... 55
      4.1.1 Electron Energy Spectrum ......................................... 55
      4.1.2 Intensity Effects ....................................................... 61
      4.1.3 Angular Distributions ................................................. 66
   4.2 Magnesium ...................................................................... 72
      4.2.1 Electron Energy Spectrum ......................................... 72
      4.2.2 Intensity Effects ....................................................... 80
      4.2.3 Angular Distributions ................................................. 87
      4.2.4 Circular Polarization .................................................. 92
   4.3 Comparisons between Calcium ....................................... 98
      and Magnesium Atoms

5. Conclusions ....................................................................... 107

References ............................................................................. 108

Appendices ............................................................................. 115
   (A) Electron Energy Analysis Program .................................. 115
   (B) Laser Intensity Effects Analysis Program ...................... 121
   (C) Angular Distribution Analysis Program ........................ 126

Vita ..................................................................................... 130
List of Tables

Table 1. Order of Nonlinearity and Saturation . . . . 65
    Intensities for Calcium.
Table 2. Atomic Parameters from Nonlinear . . . . 71
    Least-Squares Fit for Calcium.
Table 3. Order of Nonlinearity and Saturation . . . . 83
    Intensities for Magnesium.
Table 4. Comparison of Saturation Intensities . . . . 86
    for Calcium and Magnesium.
Table 5. Atomic Parameters from Nonlinear . . . . 88
    Least-Squares Fit for Magnesium.
Table 6. Comparison of the Production Ratio . . . . 101
    of Singly and Doubly Charged Ions between
    Calcium and Magnesium.
List of Figures

Fig. 1. Schematic energy diagram for sequential and direct ionization. 5
Fig. 2. Schematic representation for (a) one-photon photolization, 14
(b) nonresonant multiphoton ionization, and
(c) resonant multiphoton ionization.
Fig. 3. Schematic representation for above-threshold ionization of atoms, 16
and ATI electron spectrum of magnesium with 1064 nm radiation.
Fig. 4. Ionization of an atom in a laser field. 19
Fig. 5. Formation of singly- and doubly-charged ions. 20
Fig. 6. Schematic energy level diagram for xenon, calcium, and magnesium. 26
Fig. 7. Energy level diagram for calcium showing threshold levels relevant to this study. 28
Fig. 8. Energy level diagram for magnesium showing threshold levels relevant 33
to this study.

Fig. 9. Block diagram of the apparatus. ........................................ 39

Fig. 10. Imaging technique of measuring the laser profile. ............. 42

Fig. 11. Schematic representation of atomic beam apparatus. ......... 45

Fig. 12. Dual multichannel plate detector. .................................. 50

Fig. 13. The electron spectrum of calcium resulting from 532 nm excitation. ................................................................. 56

Fig. 14. The time-of-flight mass spectrum of calcium resulting from (a) 532 nm and (b) 1064 nm radiation ......................... 57

Fig. 15. Electron spectrum of calcium resulting from 1064 nm excitation. ................................................................. 59

Fig. 16. Log-log plots of signal versus laser intensity for (a) 532 nm and (b) 1064 nm radiation. ...................................... 62

Fig. 17. Electron angular distributions for calcium with 532 nm excitation. ................................................................. 67

Fig. 18. Electron angular distributions for calcium with 1064 nm excitation. ................................................................. 70

Fig. 19. Electron energy spectrum of magnesium with 532 nm radiation. ................................................................. 73

Fig. 20. Mass spectrum of magnesium at ................................... 75
different intensity and wavelength.

Fig. 21. Electron energy spectrum of magnesium with 532 nm radiation.

Fig. 22. Electron energy spectrum of magnesium with 1064 nm radiation.

Fig. 23. Log-log plots of signal versus laser intensity for magnesium with 532 nm.

Fig. 24. Log-log plots of signal versus laser intensity for 7-, 8-, 9-, and 10-photon ionization for magnesium at 1064 nm radiation.

Fig. 25. Electron angular distributions for magnesium with 532 nm excitation.

Fig. 26. Electron angular distributions for magnesium with 1064 nm excitation.

Fig. 27. Magnesium spectrum with circularly polarized 1064 nm radiation.

Fig. 28. Magnesium spectrum with circularly polarized 532 nm radiation.

Fig. 29. Suppression of MPI with circularly polarized light.

Fig. 30. The ionization ratio between (N)- and (N+1)-photon ionization.
ABSTRACT

When an atomic beam interacts with a high intensity laser beam, ionization occurs even if the photon energy is less than the threshold level for ionization of the atom. In this thesis, nonresonant multiphoton ionization of alkaline-earth atoms is examined experimentally. Since alkaline-earth atoms have doubly excited states just above and sometimes below the first ionization threshold level and are easily ionized with relatively low intensity, they are ideally suited for studying the role of electron correlations in multiphoton ionization. This motivated a careful and detailed investigation of the correlation of electrons and the multiphoton ionization mechanism. A spectroscopic study on photoelectrons arising from nonresonant multiphoton ionization of calcium and magnesium atoms up to $10^{13} W/cm^2$ is described. The emphasis is placed on photoelectron spectra, and their variations with laser wavelength, intensity, and polarization. Also, the ionization process of doubly charged ions which can be produced either by a stepwise process or by the simultaneous removal of two electrons is discussed.
1. INTRODUCTION

When an atom is irradiated by an intense laser beam, the atom can absorb several photons, and nonlinear phenomena and multiphoton processes occur. This multiphoton ionization (MPI) of atoms is one of the new fields of investigation in the modern physics of atoms and atomic spectroscopy that lasers have opened up over the last two decades. The observation of MPI requires strong intensities and it is only after high power lasers had been developed that the problems concerning the multiphoton interaction of atoms with a field have been studied. Some new questions arose from the MPI of atoms in strong laser fields. First, how does our view of electric dipole transitions alter in the nonperturbative regime. Second, what influence do the laser field parameters, such as wavelength, pulse width, intensity, and polarization have on our observations. Studies of multiphoton processes made possible by high-power lasers reveal new and dramatic aspects of the interaction of photons with matter, such as above threshold ionization[^1],[^2],[^3],[^4],[^5] (ATI), multiple charge state distributions[^6],[^7],[^8] (MCSD), high field Kapitza-Dirac effect[^9], short wavelength generation[^5], and strong field ionization[^10]. All of these processes can be characterized as highly nonlinear effects with the capacity for efficiently coupling large amounts of energy into an atomic system. This has practical importance for production of a laboratory X-ray or VUV laser. Although many theoretical models[^11],[^12],[^13],[^14],[^15],[^16] have been proposed to describe the salient features of these observations, the lack of experiments on open shell atoms has contributed to the inability of theorists to provide a quantitative physical picture of these effects. In this dissertation, we studied the behavior of multielectron atoms in intense laser fields which is essential to
understanding the above phenomena. Specifically, the influences of the atomic structure on the above phenomena will be stressed.

In an intense laser field, any atom undergoes substantial one-electron multiphoton ionization\textsuperscript{17,18} for intensities above $10^{9}\text{W/cm}^2$. Theoretical models in the framework of perturbation theory\textsuperscript{19,20,21,22,23}, and an independent electron approximation have been applied to describe the behavior of closed shell atoms. For intensities in the $10^{12} - 10^{15}\text{W/cm}^2$ range, the MPI of atoms displays interesting features on the electron energy spectrum. At these intensities, the interaction between a laser pulse and many-electron atoms leads to the removal of several electrons and the production of multiply charged ions\textsuperscript{24,25,26,8,27,28}. One of the simplest and the most important experiment on the MPI of multielectron atoms was done by Rhodes' group\textsuperscript{6} in 1983. They simply focused a laser beam with 193 nm wavelength and an intensity of $1 \times 10^{14}\text{W/cm}^2$ on gas-phase atoms ranging from He to U in a vacuum chamber. They found that a large number of photons were absorbed and thus a large amount of energy coupled into the system. For xenon atoms, they observed ionization up to $\text{Xe}^{6+}$ ions. Since the ionization threshold for $\text{Xe}^{6+}$ ion measured above the Xe neutral ground state is 265 eV and the photon energy for 193 nm is 6.4 eV, a 42-photon absorption is needed to produce $\text{Xe}^{6+}$ ions. According to perturbation theory, an N-photon absorption scales as $R_N = \sigma_N F^N$, where $R$ is the ionization rate, $\sigma_N$ is cross section, and $F$ is photon flux. The relative abundance for $\text{Xe}^{2+}$ to $\text{Xe}^{+}$ is $(\frac{R_{\text{Xe}^{2+}}}{R_{\text{Xe}^{+}}})T\hbar = 10^{-2}$ since $\text{Xe}^{+}$ and $\text{Xe}^{2+}$ have cross sections of $10^{-49}\text{cm}^4\text{s}$ and $10^{-115}\text{cm}^8\text{s}^3$ for a 2- and 4-photon absorption, respectively. Similarly, the relative abundance for $\text{Xe}^{6+}$ to $\text{Xe}^{+}$ is $(\frac{R_{\text{Xe}^{6+}}}{R_{\text{Xe}^{+}}})T\hbar = 10^{-10}$. However, in this experiment the rela-
tive abundance of $Xe^{2+}$ to $Xe^+$ and $Xe^{6+}$ to $Xe^+$ is $(\frac{R_{Xe^{2+}}}{R_{Xe^+}})_{Exp} = 0.6$ and $(\frac{R_{Xe^{6+}}}{R_{Xe^+}})_{Exp} = 0.02$, respectively, which are 60 and $5 \times 10^7$ times greater than those expected by perturbation theory. This extremely large abundance of highly charged states would suggest that perturbation theory is no longer valid in this intensity regime. In this experiment, multiple charge state distributions showed a strong correlation with atomic size. Specifically, atoms with small cores, that is, He, were only singly ionized, and less likely to ionize to higher charge states as compared with large core atoms, for example, Xe. However, all subsequent studies have been confined almost exclusively to rare gases, and more specifically to xenon atoms. Consequently, due to the lack of experimental data the atomic effects of these phenomena have largely been ignored. Therefore, a detailed study of these size effects is necessary in order to better understand the physics of these phenomena. Size effect studies should include not only for closed shell atoms but also open shell atoms as well. In this respect, alkaline-earth atoms are important systems to study in order to understand the influence of the atomic structure as an important parameter for MPI process. The alkaline-earth atoms with two outer shell electrons are attractive systems for MPI studies for a number of reasons. First, they contain a manifold of doubly excited states just above the first ionization threshold and in some cases even have bound doubly excited states. Thus, the density of doubly excited states around threshold for the alkaline-earth atoms is higher than for the closed shell atoms. Second, they have low ionization potential. For example, the ionization potential of calcium is 6 eV which is half of the ionization potential of xenon. Therefore, the alkaline-earth atoms are easily ionized with a lower order process, and offer the opportunity to exam-
ine these effects at moderate laser intensities, where $I < 10^{13} W/cm^2$. Finally, the low-Z alkaline-earth atoms such as calcium and magnesium are systems that should prove to be theoretically tractable. This is especially relevant considering the recent success of Kim and Greene on the single-photon ionization of calcium atoms. In order to explain the multiple charge state distributions, two mechanisms have been proposed so far (Fig.1).

First, direct ionization in which two or more electrons are removed simultaneously and the multiply charged ions are formed directly from neutral atoms’ ground state:

$$A + n\hbar\omega \rightarrow A^{q+} + qe^-.$$  

Second, sequential ionization in which the doubly charged ions are formed by a stepwise process via singly charged ions formed in the same laser radiation pulse:

$$A + n_1\hbar\omega \rightarrow A^+ + e^-,$$

$$A^+ + n_2\hbar\omega \rightarrow A^{2+} + e^-,$$

$$\vdots$$

$$A^{(q-1)+} + n_i\hbar\omega \rightarrow A^{q+} + e^-.$$  

It is clear that direct ionization implies multiple excitation. But sequential ionization may imply either single electron excitation or multiple excitation. Even if only one electron is ejected in this process, it can be regarded as multiple excitation if one electron is left in an excited state. Therefore, single electron emission may also imply multiple excitation and electron correlation should be considered. Although the extent to which multiple-electron excitation is responsible for the production of multiple-charge-state distributions (MCSD) in closed-shell atoms is still unclear, multiphoton transitions to doubly excited states have been found to play a significant role in experiments on alkaline-earths.
Fig. 1. Schematic energy diagram for sequential and direct ionization. Vertical arrows indicate photons absorbed in the photoionization process.
The production of multiply charged ions through multiphoton absorption emphasizes both atomic properties and laser characteristics such as intensity, photon energy, pulse duration, etc. In this dissertation, nonresonant MPI of alkaline-earth atoms, specially calcium and magnesium, has been studied in an intense pulsed laser field using MPI spectroscopy. We have examined the influence of atomic effects on these phenomena. That is, what role does the size of the valence and inner shell have on the effective coupling of energy into the atomic system. Also, we have studied the influence of the laser characteristics on these phenomena, such as laser intensity, photon energy, and polarization. The laser intensities of interest range from $10^{10} W/cm^2$ to $10^{13} W/cm^2$, the photon frequencies range from infrared ($h\omega = 1.16eV$) to visible ($h\omega = 2.33eV$), and a pulse duration of 10 nsec. We measured the electron energy spectrum, intensity dependence, electron angular distribution, and polarization effect for calcium and magnesium at both wavelengths in order to study the ionization process. Understanding the role of electron correlations in both single and multiphoton ionization processes is important in the area of atomic physics. Also, the mechanism of formation of doubly charged ions of alkaline-earth atoms is analyzed. We have demonstrated that sequential ionization is dominant process and electron correlations do play a significant role in single electron ionization. As to the spectroscopic aspect of these experiments, it is our objective in this work to focus on the richness of information on atomic dynamics that can be extracted. In processes of this type, however, the laser is not a simple probe, as is the case in traditional single-photon absorption. A further most desirable effect of the intensity in the present context is absorption of one or more additional
photons before a *continuum* state can decay by direct or autoionization. This is intimately connected with the possibility of leaving the ion in an excited state, as well as with double-electron ejection.

In Chapter 2, theoretical consideration for the experiment such as the structure of atoms in strong laser fields, multiphoton ionization, above threshold ionization of atoms, energy level structure of alkaline-earth atoms, and electron correlation will be dealt with. The aim of this chapter is to give a clear definitions of the concepts used here rather than to present calculations corresponding to the experimental results.

In Chapter 3, we will describe the experimental requirements for the observation of intense field effects on electrons and atoms. The laser and vacuum system, together with the various detection systems and data analysis will be presented.

In Chapter 4, the experimental results for calcium and magnesium will be presented and discussed. Electron energy and mass spectra, intensity effect, angular distribution, and circular polarization will be presented.

In Chapter 5, we conclude by summarizing the unresolved issues.
2. THEORETICAL CONSIDERATIONS

In this chapter, the theoretical basis for our experiments will be presented. We will first briefly mention the fundamentals of laser spectroscopy, and discuss the theory for the atomic structure, excitation, and ionization of atoms in strong laser fields. Then we will discuss the structure of the energy levels of calcium and magnesium which were studied in this dissertation. Finally we will discuss the electron correlations.

2.1 Multiphoton Ionization Spectroscopy

In many-electron atoms, the Coulomb repulsion and spin-orbit coupling effects between electrons strongly influence the nature of the electronic states. The one-electron picture, where each electron is viewed as moving in a well-defined orbit and independent of the motion of the other electrons, breaks down when these effects are strong enough. Multiphoton ionization spectroscopy provides us with new spectroscopic data allowing to identify previously unknown states of alkaline-earth atoms and enables us to study excited states and transitions between excited states which cannot be reached by single photon excitations. It also provides a means to probe how the Coulomb repulsion of the two outer electrons of alkaline-earth atoms\textsuperscript{35,36,37} may be taken into account.

In the technique of multiphoton ionization spectroscopy, two or more photons are absorbed simultaneously and excite atoms from the ground state to the continuum. The atomic sample is irradiated by the laser and the ions or electrons produced by laser excitation are detected by observing the pulsed cur-
rent, synchronous with the pulsed laser. Multiphoton ionization spectroscopy is based on the fact that multiphoton transitions can be induced with high probability and readily detectable with high intensity lasers. In general, the transition probability for the $N$-photon transition from $|g\rangle$ to $|f\rangle$ is,

$$W^{(N)} = \sigma^{(N)} I_1(\omega_1) \cdots I_N(\omega_1) / \hbar^N \omega_1 \cdots \omega_N$$  \hspace{2cm} (2.1)

where $\sigma^{(N)}$ is the cross section and $I_1(\omega_1)$ is the laser intensity at $\omega_1$. Then, for an $N$-photon nonresonant multiphoton ionization, the population difference produced in the excited state, $\rho_{ff} - \rho_{ff}^0$, is governed by the equation

$$\left( \frac{\partial}{\partial t} + \frac{1}{T} \right)(\rho_{ff} - \rho_{ff}^0) = W^{(N)}(\rho_{gg} - \rho_{ff}).$$  \hspace{2cm} (2.2)

where $T$ is relaxation time. In the steady state case, the population produced in the excited state is

$$\rho_{ff} - \rho_{ff}^0 = W^{(N)} T (\rho_{gg} - \rho_{ff})$$  \hspace{2cm} (2.3)

$$= \sigma^{(N)} T I_1(\omega_1) \cdots I_N(\omega_1) (\rho_{gg} - \rho_{ff}) / \hbar^N \omega_1 \cdots \omega_N.$$

From equation (2.3), excitation can be large if the cross section $\sigma^{(N)}$ and the laser intensity $I_N$ are large. The excitation is usually a complex function of the input laser intensities. With pulsed lasers, the transient response complicates the matter even more and the physics must be described using a model that includes the laser field as well as their interaction. Another interesting aspect of multiphoton ionization spectroscopy is its ability to yield Doppler-free spectra. The resolution is mainly limited by the laser line width. Since the excitation is usually weak, the detection is the most important part in MPI spectroscopy. The detection method must be extremely sensitive and it will be discussed in the next chapter.
2.2 Ionization and Structure of Atoms in a Laser Field

The ionization of an atom in a field with a frequency \( \omega \) is a multiphoton process if the energy of a quantum \( \hbar \omega \) is less than the ionization potential of the atom \( |E_o| \). This MPI can occur only if the photon flux is strong enough. The energy conservation law demands simultaneous absorption of at least \( N \) photons by the atom in a single act of interaction with the field to satisfy the relation

\[
N \geq |E_o|/\hbar \omega. \tag{2.4}
\]

The ionization process of a model system with a single level in a short-range binding potential by the field of the linearly polarized wave with a frequency \( \omega \ll |E_o|/\hbar \) was first considered by Keldysh\(^{38} \). The Keldysh method treats photoionization by considering the transition caused by the electromagnetic field on an electron from a bound state in which the Coulomb field of the ion is neglected. This method finds only the intensity parameter that arises in the high-order limit and neglects the details of the atomic structure.

In the laser-atom interactions, lowest-order perturbation theory\(^{22} \) (LOPT) for the multiphoton ionization has proved to be a powerful method in the theoretical study of various nonresonant multiphoton processes. This method treats laser fields as perturbations to atomic states and considers the process corresponding to the lowest nonvanishing order in the perturbation series as the dominant process. The basic equation of perturbative theory of MPI is given by

\[
i \hbar \frac{\partial}{\partial t} |\phi > = (-\frac{e}{m} \vec{p} \cdot \vec{A} + \frac{e^2}{2m} A^2) |\phi >. \tag{2.5}
\]

where \( |\phi > \) is constant during the time of interaction. In the frame of perturbation theory, the \( N \)-photon ionization rate \( W \) for nonresonant MPI\(^{39} \) induced by
coherent laser pulses, is given by

\[ W = \sigma_N I^N, \quad (2.6) \]

where \( \sigma_N \) is the generalized N-photon ionization cross section. \( W \) is expressed in \( s^{-1} \), \( \sigma_N \) is expressed in \( cm^{2N} s^{N-1} \) units and the laser intensity \( I \) is in \( W cm^{-2} \). What is immediately evident, is that perturbation theory predicts that the non-resonant rate scales with the intensity to the \( N^{th} \) power. This power-law behavior of the ionization probability characterizes the LOPT as a good approximation for the process and implies that the ionization process is not saturated and the process corresponding to the lowest nonvanishing order is the dominant process. Thus, a simple intensity dependent study will verify the validity of this scaling.

Fig. 2 shows schematically the N-photon ionization of an atom. The vertical arrows indicate the photons absorbed in the N-photon transition from the ground state to continuum.

The N-photon MPI requires a strong laser intensity and this can be achieved with current laser technology. In strong laser fields the perturbation is so large that \( |\phi> \) in equation (2.5) is not constant during the time of interaction and perturbation theory is not valid. Let's estimate the laser intensity for which perturbation theory breaks down. For a simple derivation of the N-photon ionization process, the single photon transition takes place with the rate,

\[ R_1 = \sigma_1(I/\hbar \omega). \quad (2.7) \]

where \( I \) is the laser intensity and \( \sigma_1 \) is the generalized one-photon cross section. The second transition must occur within a lifetime \( \tau \) of the virtual state, and the rate for the second transition is

\[ R_2 = \sigma_1(I/\hbar \omega) \tau \sigma_1(I/\hbar \omega). \quad (2.8) \]
Generalizing for \( N \) photons we get,

\[ R_N = R_N^{N-1}[\sigma_1(I/\hbar\omega)]^N, \tag{2.9} \]

where \( \sigma_N = \sigma_1^N r^{N-1} \) is \( N \) photon ionization cross section. When the MPI rate \( R_{N+1} \) approaches \( R_N \), perturbation theory is not valid any more. From equation (2.9), the breakdown occurs when \( \frac{R_{N+1}}{R_N} = r \sigma_1(I/\hbar\omega) \approx 1 \). For \( r = 10^{-15} \text{sec} \) and \( \sigma_1 = 10^{-16} \text{cm}^2 \), we get \( I/\hbar\omega = 10^{31} \text{photons/cm}^2\text{sec} \). This corresponds to \( I = 10^{12} \text{W/cm}^2 \) for 1 eV photons. For comparison, the intensity of normal daylight is \( 10^{-1} \text{W/cm}^2 \), and only \( 10^{7} \text{W/cm}^2 \) even at the surface of the sun.

Now, let’s consider the saturation intensity where the atomic population has been reduced to \( e^{-1} \) of its final value. The saturation intensity \( I_s \) for the \( N \)-photon process is

\[ I_s = \sigma^{-1/N} r^{-1/N}, \tag{2.10} \]

where \( \sigma \) is the generalized absorption cross section and \( r \) is the laser pulse duration. Here, we define

\[ \Lambda_N = (\sigma_N)^{1/N}. \tag{2.11} \]

The generalized cross section is dependent on the \( N \)-photon process and the cross section is theoretically calculated for hydrogen atom. Using the calculated hydrogenic values\(^40\) \( \Lambda_N^{(H)} \), we obtain \( \Lambda_N^{(A)} \) for an atom \( A \) through the relation\(^41\),

\[ \Lambda_N^{(A)} = \Lambda_N^{(H)} \frac{R_A^2 E^{(H)}_\infty}{R_H^2 E^{(H)}_\infty}, \tag{2.12} \]

where \( R \) represents the atomic radius, and \( E_{\infty} \) is the ionization potential. When we consider pulse duration \( r \), we get

\[ \bar{\Lambda}_N = (\sigma_N r)^{1/N} = \Lambda_N^{(A)} r^{1/N}. \tag{2.13} \]
From equations (2.10) and (2.13),
\[ I_s = \frac{1}{\Delta_N} = \Lambda_{N}^{(A)} r^{1/N}. \] (2.14)

Through equation (2.14), the saturation intensity \( I_s \) for N-photon process can be calculated.

In contrast to a nonresonant process a resonance appears in atomic processes when any excited state of the atom coincides with the energy of an integer number of photons of the intense laser field. Assume that \( N \) photons are the minimum number needed to ionize the atom. When the energy of an integer number of photons, say \( N_1 < N \) excites an intermediate resonance and the following absorption of \( N_2 = N - N_1 \) photons transfers an electron into continuum, as shown in Fig.2(c), the degree of nonlinearity determined by

\[ N(F) = \frac{\partial \log W}{\partial \log (F^2)} \] (2.15)

deviates from \( N \). Here, \( F \) is the photon flux in photons \( cm^{-2} s^{-1} \). The atom behaves like a resonator and the ionization probability can be resonantly enhanced. Such a behavior indicates that LOPT does not provide a realistic description of resonant processes and the ionization probability \( W_N \) can be formulated as

\[ W_N = \frac{2\pi |V_{E1}^{(N_2)}|^2 |V_{10}^{(N_1)}|^2}{\hbar \Delta^2 + \Gamma_0^2/4}, \] (2.16)

where \( \Gamma_0 \) is the spontaneous width and \( \Delta = E_1 - E_0 - N_1 \hbar \omega \) is the resonance detuning. \( V_{10}^{(N_1)}, V_{E1}^{(N_2)} \) are the compound matrix elements of the order of \( N_1 \) and \( N_2 \) for the transitions from the ground into the resonant state and from the resonant into the continuum state with the energy \( E = E_0 + N \hbar \omega \). Equation (2.16) shows that a resonant maximum appears in the ionization probability for
Fig. 2. Schematic representation for (a) one-photon photoionization, (b) non-resonant multiphoton ionization, and (c) resonant multiphoton ionization.
a given degree of nonlinearity $N$ and the resonance ionization probability exceeds the probability of the nonresonance process.

In a perturbative treatment, formation of singly-charged ions appears at the lowest non-vanishing order. For increasing nonlinearity, one finds the possibility for an electron to absorb more photons than the minimum number required for ionization. The electron energy of the multiphoton photoeffect is given by

$$E = Nh\omega - E_0 \quad (N = 2, 3, 4, \ldots), \quad (2.17)$$

where $N$ is the minimum number of photons of energy $h\omega$ required to ionize an atom and $E_0$ is an ionization potential. The free electrons continue to absorb photons soon after they are excited into the continuum via multiphoton absorption of this minimum number of photons required for ionization. The process of laser-induced ionization of an atom in which the electron-atom system absorbs $S$ photons more than the minimum number of photons that is required for the ionization is called above-threshold ionization\textsuperscript{43,44,1,45,46} (ATI), also called continuum-continuum transitions. The electron energy spectrum of ATI is a clear series of equally spaced peaks separated by the photon energy $h\omega$ (Fig.3). The ejected electron energy is given by

$$E = (N + S)h\omega - E_0 \quad (S = 0, 1, 2, \ldots). \quad (2.18)$$

We are interested in the final states of the system where the ion is in its ground state and the free electron has a kinetic energy given by equation (2.18). The occurrence of additional photon absorption is a normal consequence of $(N+S)$-order perturbation theory. For perturbative calculations, the ionization probabilities $P_N, P_{N+1}, \ldots, P_{N+S}$ should be calculated up to $(N+S)$-order. The maximum
Fig. 3. Schematic representation for above-threshold ionization of atoms, and ATI electron spectrum of magnesium with 1064 nm radiation.
branching ratio for the populations of the various continua is reached for the saturation intensity $I_s$, which is defined by

\[
\sum_{S=0}^{\infty} \sigma_{N+S} I^{N+S} \tau \approx 1.
\]  
\[\text{(2.19)}\]

The $(N+S)$ ionization rate$^{22,39}$ is

\[
W(N+S) = \sigma_{N+S} I^{N+S},
\]

\[\text{(2.20)}\]

where $\sigma_{N+S}$ is the generalized cross section.

Another consequence of the strong field interaction is sequential ionization in which various charge states are formed$^{47,48,49,50}$. Consider the situation where the laser beam is strong enough to eject several electrons and form doubly-charged ions in the process of MPI. Then the number of $q^+$ ions produced in the process of MPI is

\[
\sum_{q=1}^{N} N_q(t) = N_0 - N(t),
\]

\[\text{(2.21)}\]

where $N_0$ is the initial number of neutral atoms in the interaction region, $N(t)$ their number as a function of time $t$ during the pulse, and $N_q(t)$ the number of the $q^+$ ionic species. If we define $n(t) = N(t)/N_0$ and $n_q(t) = N_q(t)/N_0$ then we have the number-conservation equation,

\[
\sum_{q=1}^{N} n_q(t) = 1 - n(t).
\]

\[\text{(2.22)}\]

The time development of $n_q(t)$ is governed by a set of coupled linear differential equations from which $n(t)$ can be eliminated. The initial conditions are $n(0)=1$ and $n_q(0) = 0$. The equations are based on the transition probability $\sigma_k F^k(t)$.

Thus the rate of change of $N(t)$ is

\[
\frac{dN(t)}{dt} = - \sum_q \sigma_k F^k(t) N(t),
\]

\[\text{(2.23)}\]
where \( k_q \) is the order of the multiphoton process in which \( q \) electrons are ejected directly without going through the sequences of ions of lower charge, and \( k_q \) increases considerably with \( q \). In this notation, \( k_1 \) is the order of the multiphoton process in which one electron is ejected from the neutral atom. In this discussion, we assumed the pulse width is so short that the neutral atoms are ionized with the maximum intensity of the laser pulse (Fig.4.a). But if the pulse width is long enough the neutral atoms are ionized before it reaches the highest intensity and even saturation intensity cannot reach the maximum intensity (Fig.4.b). In this case the neutral atoms do not experience the maximum intensity of the laser pulse. Therefore, in our experiment we do not have to increase the laser intensity more than saturation intensity, because the atom can not experience that.

Now, let's consider the physical consequences of MCSD. In Fig.5(a), singly- and doubly-charged ions are shown. Singly charged \( A^+ \) ions are saturated at \( I_s \) and doubly charged \( A^{2+} \) ions are not produced at all at this intensity. In this case, the only possibility of producing \( A^{2+} \) ion is via \( A^+ \) ion. In Fig.5(b), \( A^{2+} \) ions are produced while \( A^+ \) ions are not saturated. Therefore, \( A^+ \) ions and neutral \( A \) atoms coexist at the focal point. In this case, \( A^{2+} \) ions can be made either from \( A^+ \) ions or neutral \( A \) atoms. Therefore, Fig.5(b) is the prerequisite condition for studying the direct ionization process.

In the ionization process of an atom in a laser field, one of the key questions is what happens to the ionization potential of an atom inside a laser field. In optical fields of the order of \( 10^{12} \text{W/cm}^2 \) or greater, both atoms and free electrons are fundamentally affected by the ponderomotive forces\(^{51,52} \) inherent in the strong laser light. In the realistic situation, the laser amplitude changes slowly over the
Fig. 4. Ionization of an atom in a laser field with (a) short laser pulse-width and (b) long laser pulse-width.
Fig. 5. Formation of singly- and doubly-charged ions in a laser field: (a) Singly-charged ions are saturated and no neutral atoms exist in a laser focal point. (b) Neutral atoms and singly-charged ions coexist.
electron wavelength and the laser wavelength is large compared to the electron wavelength and causes the ponderomotive potential\textsuperscript{53,54,55}. The ponderomotive potential is given by\textsuperscript{53}

\[ V(\vec{r}, t) = \frac{e^2 E^2(\vec{r}_a, t)}{4m\omega^2}, \]  

(2.24)

or in terms of intensity,

\[ V = \frac{(2\pi e^2 I)}{(mc\omega^2)}, \]  

(2.25)

where \( m \) is the electron mass, \( e \) is the electron charge, \( E(\vec{r}_a, t) \) is the electric field, \( \omega \) is the angular frequency, \( I \) is the laser intensity and \( c \) is the speed of light. The high Rydberg states and the ionization threshold of an atom in a low frequency optical field have a Stark shift\textsuperscript{56,13,57} downward given by equation (2.24), while the atom's ground state has a much smaller shift. The ionization potential of an atom is thus increased by the ponderomotive energy. Because the ionization potential is increased by the ponderomotive potential \( V \), the initial kinetic energy of the electron is reduced by this value. However, since all measurements are made outside of the beam, the ponderomotive energy converts into kinetic energy as the electron leaves the light beam, just corresponding for the decrease in the initial kinetic energy due to the raised ionization potential. Therefore, the recorded energy spectrum will not show any effects of the ponderomotive potential for slowly changing laser fields. The maximum energy the electron can gain depends only on the intensity and is given by \( E_{\text{max}}(eV) = 1.1 \times 10^{-13} I(W/cm^2)eV \) for a 1.16 eV photon energy. Therefore, the ponderomotive potential for Nd:YAG lasers at 1064 nm radiation equals the photon energy (1.16 eV) at \( 10^{13}W/cm^2 \). Ponderomotive energy shifts also affect the apparent ionization potential of the neutral atom. All energy levels in an atom are shifted to some degree by the
presence of intense laser field, because of the energy of the induced atomic po-
larization. The amount of shift depends on both the intensity and the frequency
of the field. In the case of nonresonant MPI with a Nd:YAG laser, the tightly
bound ground state is driven well below any resonant frequencies, and its polar-
izability and accompanying energy shift are therefore very small. The ionization
limit is shifted because of the ponderomotive energy imparted to the nearly free
electron, plus the much smaller induced energy of the remaining ion. Therefore,
it requires more work to ionize an atom in an intense Nd:YAG field.

Now, consider again equation (2.5). In the vicinity of the atom, the $A^2$ term
is a constant, independent of the atomic coordinates. It therefore lifts all states,
continuum and bound, by the same value, via the ponderomotive potential. If
this term were all, there would clearly be no shift in the ionization energy of the
atom. However, the term $\vec{P} \cdot \vec{A}$ describe the transition between states. If the
laser pulse is short enough, the explicit time dependence of the ponderomotive
potential causes both a shift and a broadening of the photoelectron lines in high-
intensity MPI. For high power lasers focussed into a small cross section, this
potential plays a significant role in altering the electron kinetic energy from its
value outside the laser to a lower value inside the laser. The ponderomotive
energy converts into kinetic energy as the electron leaves the laser beam. The
ionization potential of an atom is thus increased by the local ponderomotive
energy.

If some of the ponderomotive energy is lost because of the time dependence of
the ponderomotive potential, we will detect the electron at a lower energy than we
expect when the ponderomotive potential is not taken into account, i.e., $E_{\text{int}} =
where \( E_{\text{int}} \) is the initial kinetic energy of the electrons, \( E_{\text{final}} \) is the detected energy, and \( E_{\text{pond}} \) is the value of the ponderomotive potential at the intensity at which the electrons are created via ionization. From the discussion above, we should get different results for a long pulse and a short pulse, as seen by the electron. The important parameter for the criterion of pulse-length is the ratio \( d/t \), where \( d \) is the transverse diameter of the pulse at the focus and \( t \) is the pulse duration. When \( v_a \gg d/t \), where \( v_a \) is the velocity of an electron, the electron leaves the pulse so fast that the implicit time dependence that is due to the average motion of the electron dominates over the explicit time dependence due to the pulse propagation. In this case the pulse duration is so long compared with the time that it takes the electron to leave the pulse that it can be approximated by a stationary beam. There will be no energy shifts since the ponderomotive potential is approximately conserved. When \( v_a \ll d/t \) the electron does not travel any appreciable distance during the time that it interacts with the pulse. In this case the laser pulse is so short that the explicit time dependence of the ponderomotive potential dominates, and most of the energy stored in the quiver motion from the absorbed photons will be lost when the pulse separates from and leaves the electron. There will be a downward shift of the energy of the photoelectrons that is of the order of the ponderomotive potential. When \( v_a = d/t \) there will be a velocity-dependent shift. The slow electrons will be shifted more than the fast ones. The shifts are smaller than the ponderomotive potential but can still be a sizable fraction of it.
2.3 Angular Distribution of Photoelectrons

The study of photoelectron angular distributions\textsuperscript{58,10,46,59,60,61,62,63,64} for MPI gives information about the atomic structure and also tests the theories for its description. For photons having an energy less than a few electron volts, the transfer of the photon linear momentum can be neglected. In this approximation the angular distribution of photoelectrons in one-photon ionization of ground-state atoms by linearly polarized light is given by the differential cross section for photoionization as\textsuperscript{65,60,67}

\[ \frac{d\sigma^{(1)}}{d\Omega} = \frac{\sigma_{\text{tot}}}{4\pi} [1 + \beta_2 P_2(\cos \theta)], \quad (2.26) \]

where \( \theta \) is the angle between the direction of the outgoing electron and the photon polarization; \( P_2 = \frac{1}{2}(3\cos^2 \theta - 1) \) is the second Legendre polynomial; \( \sigma \) is the angle integrated (total) photoionization cross section; and \( \beta_2 \) is the angular distribution parameter which varies depending on the specific process under consideration. For unpolarized light\textsuperscript{65}, \( \theta \) is the angle between the directions of outgoing electron and light propagation, and the parameter \( \beta \) is replaced by \( -\frac{1}{2} \beta \) in equation (2.26). This equation for the angular distribution reflects the dipole character of the interaction between the photon and the atom. For N-photon ionization of an atom, a sensitive and important feature of any MPI process is the photoelectron angular distribution. For nonresonant frequencies, the angular distribution is determined by a superposition of these channels with relative weights determined by the respective matrix elements and Clebsh-Gordon coefficients. The generalized cross section \( p(\theta) \) can be written as

\[ p(\theta) = p_0 + p_2 \cos^2 \theta + p_4 \cos^4 \theta + \cdots, \quad (2.27) \]
where the time-independent coefficients $p_k$ depend only on atomic parameters and not on laser intensity. The light is linearly polarized and $\theta$ is the angle between the polarization vector and the propagation direction of the photoelectron. Regarding $p(\theta)$ as the probability of ejecting an electron at an angle $\theta$ with the direction of polarization, the distribution of the direction of flight of photoelectrons can be described as

$$
\frac{d\sigma(N)}{d\Omega} = \frac{\sigma_{tot}^{(N)}}{4\pi} \sum_{j=0}^{N} \beta_{2j} P_{2j}(\cos \theta)
$$

(2.28)

and

$$
I(\theta) \propto \sum_{j=0}^{N} \beta_{2j} P_{2j}(\cos \theta),
$$

(2.29)

where $N$ is the number of photons absorbed by each atom, $\sigma_{tot}^{(N)}$ is the total cross section for N-photon ionization, $P_{2i}(\cos \theta)$ is the Legendre polynomial of order $2i$, and $\beta_{2i}$ is an angular distribution parameter which contains the matrix elements. The physical interpretation of this equation is that the original system consists of the spatially isotropic atom and the incoming photon. Since the interaction of the photon and the atom has dipole character, the total system has dipole anisotropy, which is also displayed in the photoelectron angular distribution.

2.4 Energy Level Structure of Alkaline-Earth Atoms

Figure 6 contrasts the differences between xenon and two alkaline-earth atoms, as well as the differences in atomic structure between calcium and magnesium. One of the general aims of this dissertation is to emphasize the importance of the atomic structure on the physics of the atom - strong field interaction.
Fig. 6. Schematic energy level diagram for xenon, calcium, and magnesium.

Vertical arrows indicate photons absorbed in the photoionization process.
Compare the structure of xenon, or any inert gas, to that of an alkaline-earth atom. First, and most obvious, is that the ionization potential of both calcium and magnesium are approximately half that of xenon. Therefore, only moderate laser intensities are needed to study nonresonant ionization. Second, the alkaline-earth atoms are well suited for studying the role of electron correlations in nonresonant MPI. This is a consequence of low lying threshold levels which give rise to a high density of doubly excited states (autoionizing Rydberg series) near the first ionization potential. Third, the alkaline-earth atoms will pose a much simpler and important model system for future theoretical studies. The difference between calcium and magnesium is more subtle but note that the $n d$ threshold in calcium is very near threshold while in magnesium it is $9 \text{ eV}$ higher. In fact, the alkaline-earth atoms can be categorized into two different groups, the low Z Mg and Be atoms, and the higher Z, Ca, Sr, and Ba atoms.$^{68}$ In this chapter, we review some basic features of the alkaline-earth atoms, with special emphasis on calcium and magnesium.

2.4.1 Energy Levels of Calcium Atoms

Calcium is an alkaline-earth element ($Z=20$) with ground state configuration

$$4^1 S_0 (1s^2 2s^2 2p^6 3s^2 3p^6 4s^2).$$

The energy level diagram, shown in Fig.7, is characteristic of the alkaline-earth elements. The kinetic energy of the ejected electron is given in the parentheses and the equation number correlates with the path. The ionization threshold values for the ground state of the neutral and singly ionized calcium are 6.11 and
Fig. 7. Energy level diagram for calcium showing threshold levels relevant to this study. The numbers in parenthesis correlate with the equations given in the text. The energies for the threshold levels are referenced with respect to the neutral calcium 4s$^2$ ground state.
$11.87 \text{ eV}$, respectively. The lowest-order ionization process for neutral ground-state calcium with 532 nm radiation is

$$Ca(4s^2) + 3h\nu \rightarrow Ca^+(4s) + e(0.88\text{eV}). \quad (1)$$

Similarly for 1064 nm radiation,

$$Ca(4s^2) + 6h\nu \rightarrow Ca^+(4s) + e(0.88\text{eV}). \quad (2)$$

Sequential ionization results in the creation of doubly ionized calcium by subsequent excitation of the ground state ions created in the first two paths. For lowest-order ionization with 532 nm radiation,

$$Ca^+(4s) + 6h\nu \rightarrow Ca^{2+}(3p^6) + e(2.11\text{eV}); \quad (3)$$

while for 1064 nm excitation,

$$Ca^+(4s) + 11h\nu \rightarrow Ca^{2+}(3p^6) + e(0.95\text{eV}). \quad (4)$$

All of the above multiphoton processes are adequately described by (a) a model incorporating the transition dynamics for single-electron ionization, that is, with each process resulting in a free electron and a ground-state ion and (b) the dynamics of the laser pulse.

A sharp deviation from this simple single-electron picture is direct double ionization of neutral calcium above the two electron escape limit. The result of this will be the simultaneous ejection of two correlated electrons. An 8-photon absorption of 532 nm radiation yields

$$Ca(4s^2) + 8h\nu \rightarrow Ca^{2+}(3p^6) + 2e(\text{continuous}); \quad (5)$$
while 1064 nm photon absorption proceeds via

\[ \text{Ca}(4s^2) + 16h\nu \rightarrow \text{Ca}^{2+}(3p^6) + 2e(\text{continuous}). \]  

(6)

The signature for this process will be distinct from path (1) through (4). First and most obvious, the order of nonlinearity is higher for paths (5) and (6) than for any of the individual sequential processes. Second, instead of discrete electron peaks, one would expect to observe a flat continuous spectrum from zero energy up to the maximum kinetic energy. This maximum kinetic energy is determined by the excess energy of the \( N^{th} \) photon absorption above the two-electron threshold. The simple physical interpretation is that all combinations for sharing the excess energy between the two electrons are equally probable. Furthermore, a closer theoretical examination for single-photon absorption reveals a slight preference or cusp in the distribution at half the maximum kinetic energy. A number of groups have looked for explicit evidence for direct ionization with no satisfactory results. Although the spectral and angular earmark for this process is quite distinct from sequential ionization, practical experimental difficulties at these intensities render detection of direct ionization difficult because of contributions from background electrons.

Above-threshold ionization has been studied extensively in the rare gases by various groups and the results are satisfactorily described in terms of the one-electron model. This is particularly due to the fact that in the rare gases no multiply excited states exist close to threshold. However for calcium, as well as other alkaline-earth atoms, additional photon absorption can result in a physical picture quite different from single-electron ATI. This is a consequence of the atomic structure of calcium where doubly-excited state manifolds are found
close to threshold and represent additional open decay channels. This is seen in Fig.7, where the 3d and 4p thresholds lie approximately 1.6 and 3.1 eV above the 4s limit. In addition, some of the doubly excited states are bound in the neutral, i.e., $4p^2$, $3d^2$ states. Consequently, there is an enhanced probability that the multiphoton process will acquire some double-excitation character and possibly lead to an observable branching ratio for direct two-electron ionization. Specifically, if an additional fourth 532 nm photon is absorbed in the continuum the excitation results in three open decay channels:

\[
Ca(4s^2) + (3 + 1)\nu \begin{cases} 
  \rightarrow Ca^+(4s) + e(3.21eV) & \text{ATI for 0.88 eV} \\
  \rightarrow Ca^+(3d) + e(1.51eV) & \text{(8)} \\
  \rightarrow Ca^+(4p) + e(0.07eV) & \text{(9)}
\end{cases}
\]

Path (7) is an electron decay channel which results in a spectrum similar to that observed in the ATI of rare-gas atoms. The kinetic energy of the electron in path (7) differs from the electron energy in path (1) by the photon energy $\nu$. However, path (8) and (9) leave the ion in an excited 3d metastable state and excited 4p state, respectively. The salient point is that the final state of the process results in two excited electrons and consequently the single-electron model breaks down. Likewise, path (1) and (7) would have also evolved with some probability of two-electron correlation. Thus, we see a clear distinction could exist between the mechanism responsible for ATI in rare gases and alkaline-earth atoms. In a similar manner, for 1064 nm excitation the absorption of one and two additional photons results in

\[
Ca(4s^2) + (6 + 1)\nu \begin{cases} 
  \rightarrow Ca^+(4s) + e(2.04eV) & \text{ATI for 0.88 eV} \\
  \rightarrow Ca^+(3d) + e(0.35eV) & \text{(10)}
\end{cases}
\]

and

\[
Ca(4s^2) + (6 + 2)\nu \begin{cases} 
  \rightarrow Ca^+(4s) + e(3.21eV) & \text{ATI for 0.35 eV} \\
  \rightarrow Ca^+(3d) + e(1.51eV) & \text{ATI for 0.35 eV} \\
  \rightarrow Ca^+(4p) + e(0.07eV) & \text{(14)}
\end{cases}
\]
The (6+1) process leads to two open decay channels, 4s and 3d, and the (6+2) process leads to the addition of the 4p decay channel. There are many other higher-order processes that could be included in this discussion, e.g., ionization of Ca$^+$ from an excited initial state. We limit this discussion only to those processes that are currently observable with our detection sensitivity but do not exclude the others as relevant mechanisms.

2.4.2 Energy Levels of Magnesium Atoms

The ground state configuration of magnesium (Z=12) is

$$3^1S_0(1s^22s^22p^63s^2).$$

The energy level diagram is shown in Fig.8. The ionization threshold values for the ground state of the neutral and singly ionized magnesium are 7.64 and 15.04 eV, respectively. The lowest-order ionization process for neutral ground-state magnesium with 532 nm radiation is

$$Mg(3s^2) + 4h\nu \rightarrow Mg^+(3s) + e(1.68eV). \quad (1)$$

Similarly for 1064 nm radiation,

$$Mg(3s^2) + 7h\nu \rightarrow Mg^+(3s) + e(0.51eV) \quad (2)$$

The doubly ionized magnesium by sequential ionization with 532 nm radiation is,

$$Mg^+(3s) + 7h\nu \rightarrow Mg^{2+}(2p^6) + e(1.28eV); \quad (3)$$

while for 1064 nm excitation,

$$Mg^+(3s) + 13h\nu \rightarrow Mg^{2+}(2p^6) + e(0.11eV). \quad (4)$$
Fig. 8. Energy level diagram for magnesium showing threshold levels relevant to this study. The numbers in parenthesis correlate with the equations given in the text. The energies for the threshold levels are referenced with respect to the neutral magnesium 3s2 ground state.
For direct double ionization, a 10-photon absorption of 532 nm radiation yields,

\[ Mg(3s^2) + 10h\nu \rightarrow Mg^{2+}(2p^6) + 2e(\text{continuous}); \]  

(5)

while 20 photon absorption of 1064 nm proceeds via

\[ Mg(3s^2) + 20h\nu \rightarrow Mg^{2+}(2p^6) + 2e(\text{continuous}); \]  

(6)

For magnesium, as well as calcium atoms, additional photon absorption results in ATI. For 532 nm radiation the absorption of one or more additional photons results in following open decay channels:

\[ Mg(3s^2) + (4 + 1)h\nu \rightarrow Mg^+(3s) + e(4.01\text{eV}) \text{ ATI for 1.68eV} \]  

(7)

\[ Mg(3s^2) + (4 + 2)h\nu \begin{cases} \rightarrow Mg^+(3p) + e(1.91\text{eV}) \\ \rightarrow Mg^+(3s) + e(6.35\text{eV}) \end{cases} \text{ ATI for 1.68 eV} \]  

(8)

(9)

\[ Mg(3s^2) + (4 + 3)h\nu \begin{cases} \rightarrow Mg^+(4s) + e(0.01\text{eV}) \\ \rightarrow Mg^+(3p) + e(4.25\text{eV}) \text{ ATI for 1.91 eV} \\ \rightarrow Mg^+(3s) + e(8.68\text{eV}) \text{ ATI for 1.68 eV} \end{cases} \]  

(10)

(11)

(12)

Path (7), (9), and (12) show above threshold ionization for 3s state which differ from the electron energy in path (1) by the multiples of the photon energy \( h\nu \). Also, additional photon absorption results in excited states. Path (8) and (11) leave the ion in an excited 3p metastable state, and the kinetic energy of the electron in path (11) differs from the electron energy in path (8) by the photon energy \( h\nu \). Path (10) leaves the ion in an excited 4s state.

In a similar manner, for 1064 nm excitation the absorption of one and more additional photons results in

\[ Mg(3s^2) + (7 + 1)h\nu \rightarrow Mg^+(3s) + e(1.68\text{eV}) \text{ ATI for 0.51eV} \]  

(13)
\[
Mg(3s^2) + (7 + 2)h\nu \rightarrow Mg^+(3s) + e(2.84eV) \quad \text{ATI for 0.51eV} \quad (14)
\]
\[
Mg(3s^2) + (7 + 3)h\nu \rightarrow Mg^+(3s) + e(4.01eV) \quad \text{ATI for 0.51eV} \quad (15)
\]
\[
Mg(3s^2) + (7 + 4)h\nu \left\{ \begin{array}{l}
\rightarrow Mg^+(3p) + e(0.75eV) \\
\rightarrow Mg^+(3s) + e(5.17eV)
\end{array} \right. \quad \text{ATI for 0.51 eV} \quad (16)
\]

Since \(3p\) and \(4s\) thresholds lie approximately 4.4 and 8.7 eV above the \(3s\) limit, the \((7+1)\) process through \((7+3)\) process leads only to ATI for \(3s\). The \(3p\) excited decay channel starts to appear in the \((7+4)\) process. Therefore, the ATI peaks will be dominant for Mg with 1064 nm radiation which is different to calcium atom. Another difference between Ca and Mg for our experimental situation is that the bound \(3s5d\) intermediate state in Mg is near resonance with our photon energy. This intermediate state may act as a resonance with 1064 nm radiation.

2.5 Correlations of Two Excited Electrons

In case of alkaline-earth atoms, there are two valence electrons. These electrons may be simultaneously excited in the strong laser field and consequently manifest some degree of electron correlation. The correlation of two excited electrons can be expressed as Hamiltonian:

\[
H = -\nabla_1^2 - \nabla_2^2 + V(r_1) + V(r_2) + \frac{1}{r_{12}},
\]

where \(r_1\) and \(r_2\) are the radial positions of the two electrons and \(r_{12}\) is their separation. \(V(r_1) = -(2/r_1)\) and \(V(r_2) = -(2/r_2)\) for a \(Ca^+\) or \(Mg^+\) ion, and \(\sum_{l=0}^{\infty} \frac{r_1^l}{r_2^{l+1}} P_l(cos\theta_{12})\) for \(r_1 < r_2\).

\[
H = -\nabla_1^2 - \nabla_2^2 - \frac{2}{r_1} - \frac{2}{r_2} + \sum_{l=0}^{\infty} \frac{r_1^l}{r_2^{l+1}} P_l(cos\theta_{12}).
\]
If we ignore the higher order term, the solution separates into a product of two single-particle wavefunctions. In this approximation, the wavefunction for the final doubly excited state $nlnl'$ is

$$\psi(nln'l') = \psi(nl)\psi(n'l'),$$

(2.32)

where $n$ and $n'$ are the principal quantum numbers for inner electron, and $l$ and $l'$ are the orbital angular momentum quantum numbers. Here, the configuration interaction is allowed only by including higher order terms. Therefore, the interaction of an electron with any single one is very weak as long as the electrons are constrained to closed shells. However, when both electrons are excited outside the valence shell, the interaction between the two electrons has a major influence. Also, a pair of electrons in the field of an ionic core belongs to an open shell even in its states of lowest energy. This pair displays significant correlations even while it remains in the ground state shell\textsuperscript{72}. Electron correlations play an important role in determining the structure of doubly excited states, and this effect is especially strong in alkaline-earth atoms.

In case of calcium with a $4s^2$ ground state, it has $4p$ orbitals available for excitation within the valence shell as well as $3d$ orbitals. Photoexcitation of a calcium atom\textsuperscript{68} thus leads not only to the normal excitation $4sn'p$ or to ionized states $4sep$ but also to doubly excited states $4pn's$, $4pces$, $4pn'd$, $4ped$, $3dn'p$, and $3dep$ configurations which are extensively mixed by correlations. Since the $3d4p$ level (4.44 eV) of calcium lies well below the Ca$^+$ ion threshold (6.11 eV), the strong electron correlation with equal amplitudes of $4snp$ and $3dnp$ effectively embeds the $3d4p$ level into a sequence of $4snp$ levels, distributing its strong intensity among many other lines\textsuperscript{73}. Therefore, the influence of doubly excited
states of Ca following multiphoton excitation may appear in the electron energy spectrum. A clear signature could be the existence of electron peaks that decay to the 3d and 4p excited levels of Ca\(^+\) ion. The 3d and 4p excited states will appear in our spectra at 1.51 eV and 0.07 eV with 532 nm excitation, and 0.35 eV and 0.07 eV with 1064 nm excitation.

The spectrum of magnesium is a little different from that of calcium. The main difference between Mg and Ca lies in the replacement of the excited \((n - 1)\)de\(P^o\) channel by a higher energy channel \(n\)d\(e\)s\(P^o\). For magnesium, the \(P^o\) potential curve leading to the Mg\(^+\)(3d) threshold is highly excited. Even the \(P^o\) curve leading to the Mg\(^+\)(3p) lies further up from the potential curve leading to Mg\(^+\)(3s). Correlation effects are thus less apparent in the spectrum of magnesium than they are in calcium. On the other hand, the \(P^o\) autoionization spectrum of magnesium shows strong electron correlations. In the electron energy spectrum of magnesium, the doubly excited state 3p will appear at 1.91 eV and 0.75 eV with 532 nm and 1064 nm, respectively. The common feature of these alkaline-earth atoms is the strong correlation of the excited electrons thereby requiring small energy to release two electrons from the shell. Therefore, only moderate laser intensity (\(\sim 10^{12} W/cm^2\)) is necessary to study the correlation of the excited electrons of Ca or Mg atoms.
3. EXPERIMENTAL SETUP

In this work we have studied nonresonant multiphoton ionization in an intense long wavelength laser field. A block diagram of the experimental apparatus is illustrated in Fig.9. Neutral atoms, continuously produced in an evacuated oven, are excited and ionized by optical pulses of short duration from a Nd:YAG laser. The laser and atomic beams intersect at right angles in the high vacuum chamber. Either electrons ejected from the atom or ions created in the process are analysed with a time-of-flight (TOF) spectrometer. The arrival times of the electrons or ions to our detector are recorded by a 200 MHz transient recorder and subsequently stored in a PDP 11/73 computer. The data is analyzed for electron energies and fragment mass-to-charge ratio, intensity dependence, and angular distributions of electrons. The laser is operated at the same constant energy output and the power is externally attenuated. A KDP nonlinear crystal is used when the second harmonic radiation at 532 nm is employed. For the intensity studies, the polarization of the laser is parallel to the direction of TOF spectrometer. For angular distribution studies, the polarization of the laser is rotated by means of a half-wave plate. Some part of the laser beam is reflected by a beam splitter to a photodiode and this analogue signal from photodiode is used to trigger the transient recorder and provide a constant monitor of the laser power.

3.1 The Laser and the Optical System

Since the basic principle of this experiment is to understand the interaction
Fig. 9. Block diagram of the apparatus, described in text. The power attenuator consists of a half-wave plate, W1, and polarizer, P. The harmonic generator, SHG, is used only in the 532nm studies. The polarization of the laser is rotated by means of a half-wave plate, W2. A beam splitter, BS, reflects part of the laser beam to a photodiode, PD. The analog signal from PD is used to trigger the transient recorder and provide a constant monitor of the laser energy. The laser and atomic beams intersect at right angles and is only shown in the figure as collinear for illustrative convenience.
between a beam of laser light and atoms, a critical concern in this experiment is the control and knowledge of the laser. The important parameters for the laser are the mode structure, interaction volume, and intensity distribution, and these should be kept constant during the experiment in order to get meaningful data. We use a Nd:YAG (yttrium aluminum garnet) laser. The oscillator consists of a Nd doped YAG crystal rod 120 mm long and 7.0 mm in diameter which is pumped by two flash lamps. The laser is Q-switched by a Pockels cell - quarter wave plate arrangement. The oscillator output consists of a 500 mJ pulse of 1064 nm light with a duration 10 ns (FWHM). After a 0.5 m path, the laser pulse is amplified to an energy of 1.5 J by an additional 120 mm long, 9 mm diameter Nd:YAG rod. The laser radiation output is linearly polarized. The laser wavelength is centered at 1064 nm, which corresponds to a frequency of 9398.5cm⁻¹. The second harmonic at 532 nm (18797cm⁻¹) is generated when needed by using a KDP crystal with a conversion efficiency of 35 %. The light has linear polarization parallel to the axis of the electron spectrometer. We shielded all cables and the YAG laser in a RF (radio frequency) isolated, grounded enclosure to minimize RF interferences.

The mode structure of the YAG laser depends upon the details of the optical resonator and nonlinear effects of the cavity material. In the Nd:YAG laser, thermal lensing is known to have a significant effect on the mode quality of the output. Thermal lensing is a self focusing effect caused by the nonlocal character of the nonlinearity which results from thermoconductivity. It leads to a different rate of field enhancement in the focus, and causes spherical aberration for a Gaussian beam. Since the thermal lensing is controlled by the accumulated
energy, we must operate the laser at constant thermal load in order to keep the mode structure constant. When we need to change the laser intensity we attenuate the fixed energy output of the laser with external optics consisting of a half-wave plate followed by a Brewster angle polarizer. Rotation of the laser polarization with respect to the detector is accomplished with a half-wave plate. The retardation plates are aligned using independent checks to minimize any asymmetries caused by imperfect optics. After manipulating the intensity and polarization, the laser pulse is focussed into a vacuum chamber by a 100mm focal length lens. We measured the focus of our 100 mm focal length lens in order to understand the energy distribution at the focal waist. The highly nonlinear physical process of MPI dictates a precise knowledge of the laser intensity in the focus. The measurement of the laser profile in the focus for the YAG fundamental (1064 nm) and second harmonic (532 nm) wavelengths is accomplished by imaging techniques (Fig.10). The focus is imaged with a lens and the image positioned in the far field. The magnification of the lens is calculated with the help of geometrical optics. A slit is moved through the magnified focus and the transmitted beam energy is measured as a function of its position. The derivative of the curve yields the information about the spatial beam profile. The width which corresponds to the $e^{-1}$ of the maximum intensity is measured as the diameter of the focus. For the 1064 nm laser beam, the diameter of the focus is measured to be $50 \pm 5 \mu m$. The maximum pulse energy $F$ at which spectra are taken is 1500 mJ, corresponding to an average intensity $I$ in the focus

$$I_{max} = \frac{F}{(\pi/4) \lambda^2} \approx 1 \times 10^{13} W/cm^2. \quad (3.1)$$

For the 532 nm laser beam, the diameter of the focus is $25 \pm 3 \mu m$ and the
Fig. 10. Imaging technique for measuring the laser profile. The diameter of a laser focal point with lens $a$ is $d/m$ where $m$ is the magnification ratio of lens $b$. 
maximum intensity taken at 400 mJ is again $1 \times 10^{13} \text{W/cm}^2$. Our absolute intensity is known with an accuracy of 30\% since the diameter of the focus has an error of 10\%. The laser energy is constantly monitored during a data run and the shot-to-shot fluctuations are kept to ±1\%. We align the YAG laser focusing lens for very carefully with He-Ne laser. First, the He-Ne laser is aligned to counter-propagate with the YAG laser beam. Next, the He-Ne laser beam is allowed to pass through the center of the holes on the $\mu$-metal shield of the electron spectrometer. Two apertures are placed outside the chamber so that they are on line with the two holes on the $\mu$-metal shield. After that, we place the 100 mm focal length lens on the side of incoming laser beam. The focal point is aligned to lie at center of the electron spectrometer. The incoming and outgoing laser beams are carefully guided through the holes on the $\mu$-metal shield to avoid any scattering effects. The lens is oriented so that it has maximum intensity at focal point by observing the threshold value for air breakdown.

3.2 Vacuum System

The vacuum is a differentially pumped atomic beam apparatus. The atomic source chamber has an effusive oven and is evacuated by a liquid-$N_2$-trapped 6-inch diffusion pump. The cold trap provides sufficient protection against diffusion pump oil backstreaming. The source chamber achieves an ultimate pressure of $10^{-8}\text{torr}$. A 1mm skimmer separates the source chamber from the interaction-region chamber. The interaction region is surrounded by a 3-inch cylindrical conetic magnetic shield which also provides shielding for the drift region of the electron spectrometer. The laser and atomic beams intersect at right angles in
the shielded region via four axial holes in the shield. Located outside the shielded region is a similar 6-inch diffusion pump system to handle the load of the atomic beam. This region of the chamber is maintained at $10^{-8}$ torr. In order to further reduce the pressure in the interaction region, a 360 ℓ/sec turbomolecular pump independently evacuates the inner volume of the cylindrical shield. This method achieves a pressure in the interaction region of $10^{-9}$ torr. This is especially crucial since all the experiments are performed using low atomic-beam densities in order to minimize any effects caused by space charge and uncontrolled background of ions and electrons.

The experiments were done in a thermal atomic beam produced in the apparatus depicted in Fig.11. Chemical grade magnesium or calcium is placed in a molybdenum crucible which sits in a ceramic oven. The crucible is evacuated to $10^{-8}$ torr and baked 72 hours before doing the experiment. For the experiment, the crucible is heated with tungsten heating filaments which are powered by line voltage controlled by a variac. The oven is operated at 400 to 450°C for magnesium and 500 to 550°C for calcium. The vapor pressures of magnesium and calcium deduced from the vapor-pressure tables are about $10^{-5}$ torr. Using the equation of state of an ideal gas $PV = \frac{1}{3} N \bar{m} \bar{v}^2$, we get an atomic number density of $1.3 \times 10^{17} m^{-3}$ in the oven, and $6 \times 10^{15} m^{-3}$ at the interaction region 40 cm downstream from the oven. The volume of the laser at the focal point is $5 \times 10^{-14} m^3$ for 1064 nm radiation and $6 \times 10^{-15} m^3$ for 532 nm radiation. Assuming that all the atoms are ionized, the total number of ions produced at the focal point is 100/shot for 1064 nm and 15/shot for 532 nm radiation. The skimmer in front of the oven serves to collimate the beam so that it interacts
Fig. 11. Schematic representation of atomic beam apparatus.
with the laser beam.

### 3.3 Time of Flight Spectrometer

The time-of-flight (TOF) spectrometer is a 31.5 cm long, field free volume with an electron detector at the end. When electrons are ejected after a multiphoton process they diffuse freely out of the interaction volume with energies from a fraction of eV to a few tens of eV. The energy spectrum can be determined by measuring the arrival time of electron with respect to the initial laser pulse over a known flight length. Also, when the direction of the light field polarization is scanned, the angular distribution is obtained. In experiments on MPI of atoms, an intense pulse of laser is focussed onto a very small region of an atomic beam. The kinetic energy of the emitted electron is characterized by its mass $m_e$ and velocity $v$ by the simple relationship,

$$E = \frac{1}{2} m_e v^2$$

$$E = \frac{1}{2} m_e \frac{l^2}{t^2}$$

where $l$ is the length of the TOF tube and $t$ is the flight time for the electron to reach the detector. Therefore, the flight time becomes

$$t = l \sqrt{m_e/2E}$$

$$t = \frac{l}{\sqrt{2}} \sqrt{m_e/E}$$

$$t = 1.69 \times 10^{-8} l (\text{cm})/\sqrt{eV} \quad (\text{sec})$$

$$t = 16.9 l (\text{cm})/\sqrt{eV} \quad (\text{nsec})$$

The error associated with the uncertainties in the spectrometer is

$$\left( \frac{\Delta E}{E} \right)_{TOF} = \left[ \left( \frac{2\Delta t}{t} \right)^2 + \left( \frac{2\Delta l}{l} \right)^2 \right]^{1/2},$$
where $\Delta t$ is the time resolution of the apparatus and $\Delta l$ is the electron flight path uncertainty.

Our time-of-flight (TOF) spectrometer consists of a 2.5 cm diameter, 31.5 cm long, magnetically shielded copper Faraday cage. A thin molybdenum circular plate with a 5mm-diam hole bored at its center is placed at the beginning of the flight tube and acts as the entrance aperture of the spectrometer with the acceptance angle $3^\circ$. This plate provides both a defining aperture for the spectrometer which minimizes stray background counts and a means of providing further differential pumping of the TOF spectrometer with a $55\ell/sec$ turbomolecular pump. The interaction region and TOF electron analyzer are shielded from stray electric and magnetic fields with $\mu$ - metal. In the present experiment, electrons of energy $0 - 10eV$ formed in MPI were collected with this spectrometer. Since the diameter of the focus is $25\mu m$, the uncertainty in the electron flight path is $\Delta l = 2.5 \times 10^{-3}l$. The pulse length of the laser is 10 ns, time discrimination of the transient digitizer is 5 ns, and the total time resolution is 15 ns. For these parameters, the flight time of the electron for $0.5 - 5eV$ is

$$t = \frac{(16.9)(31.5)}{\sqrt{0.5 - 5.0}} ns = 240 - 760ns,$$

and the uncertainty of the energy is

$$\left(\frac{\Delta E}{E}\right)_{TOF} = \left[\left(\frac{2 \times 15}{760 - 240}\right)^2 + (2 \times 2.5 \times 10^{-3})^2\right]^{\frac{1}{2}},$$

$$\approx 0.4\% - 1.3\%$$

and the main uncertainty is determined by the flight time. The energy resolution is 16 meV at 0.5 eV and 65 meV at 5 eV. Our resolution is approximately a factor of two worse than this result. This is probably a consequence of stray fields
and varying contact potentials. Appreciable space charge distribution results in
the broadening of the spectrum via a Coulomb repulsion. Consequently, space
charge broadening is also a factor which can degrade the resolution. This can
easily be avoided by operating at low atomic beam density or laser intensity. We
reduced this systematically by varying the atomic beam density until no spec-
tral broadening was observed in the electron energy spectrum. For our intensity
studies, we varied the laser intensity and maintained the atomic beam density
low enough to avoid space charge effect at high intensity regime. For angular
distribution measurements, the laser intensity was kept constant and at a value
less than the saturation intensity. Another factor which degrades the resolution
and electron energy spectrum shift is the deposition of alkaline-earth atoms on
the spectrometer. Since alkaline-earth atoms are chemically reactive, the atoms
condense on cold surface with time. We tried to minimize the scattering by
trapping stray alkaline-earth atoms with a liquid nitrogen cold trap close to the
interaction region. However, some of them will scatter and stick to electron spec-
trometer and cause time dependent energy shifts via varying contact potentials.
In order to get reliable data, we disassembled the spectrometer and cleaned it
throughly with methanol after a 24 hour data collection period. The inside of the
flight tube and the molybdenum plate which is at the end of the spectrometer
are sprayed with aerodag. Aerodag is a dry-film lubricant of specially processed
micro-sized graphite in isopropyl alcohol. While applying the aerodag we heat
up the flight tube in order to yield a uniform film of aerodag on the surface.
Aerodag improved the resolution of spectrometer by reducing patch effect which
is caused by the differences in contact material potentials. In order to verify
the calibration of the electron spectrometer, we have calibrated the spectrometer with xenon. We measured electron energy spectrum of xenon and compared with the previously reported values for each of the peaks. For calibration, we observed \( P_{1/2} \) and \( P_{3/2} \) peaks of xenon which are at 0.54 eV and 1.85 eV with 532 nm excitation, and 0.54 eV and 0.68 eV with 1064 nm excitation, respectively.

The time-of-flight spectrometer has also been used in analysing the created ions. The ions are extracted and repelled by a D.C. electric field through a hole, and fall on a detector. The basic equation for the motion of ions is

\[
qE = \frac{1}{2} \frac{m}{t^2}. \tag{3.7}
\]

The flight time for the \( Mg^+ \) and \( Mg^{2+} \) ions are

\[
t_{Mg^+} = \sqrt{\frac{m}{2qE}} x = \sqrt{\frac{24}{2 \cdot 1 \cdot E}} x, \tag{3.8}
\]

and

\[
t_{Mg^{2+}} = \sqrt{\frac{24}{2 \cdot 2 \cdot E}} x = \frac{1}{\sqrt{2}} t_{Mg^+}. \tag{3.9}
\]

Therefore, we can find out a mass spectrum of multiply charged ions from singly charged ions by measuring the relative time of arrival.

3.4 Electron and Ion Detection System

A modified dual microchannel plate (MCP) detector (Fig. 12) is used to detect photoelectrons or positive ions. Microchannel plates have direct sensitivity to charged particles and energetic photons. In our detector, the plates are separated by 80\( \mu m \) and individually operated at gains in the 10^4 range. The bias voltage is
Fig. 12. Dual multichannel plate detector: (a) electron detector and (b) ion detector.
kept at 2.2 kV and the output electrons are accelerated by 200 V to a collecting plate. Three thousandth of an inch thick mylar is inserted between flat gold deposited plate and a 50Ω impedance matched aluminum conical anode. This scheme results in capacitively decoupling the output from the high voltage while maintaining good temporal response. Also, 1MΩ – 1pf high frequency filters are added to all high voltage lines to remove ringing in the output. An output pulse at 10^7 gain develops about 100 mV signal across a 50Ω load. The detector at the end of the drift tube provides an angular resolution of 2 × 10^{-3}sr with respect to the source. The signal is fed to a 200 MHz amplifier and then into a 200 MHz discriminator. Care is taken to ensure that no pulse pile-up occurs and that the statistics are representative of single-electron counts. The discriminated output is then fed into a 200 MHz transient recorder. The transient recorder is interfaced to a PDP 11/73 computer via a CAMAC acquisition system. The electron spectrum is recorded for every laser shot. The maximum total ion yield per laser shot is limited to 50 - 100 ions. This reduces approximately to one electron count per shot detected through the spectrometer’s acceptance angle when the laser’s polarization is parallel to the detector’s axis.

3.5 Data Acquisition and Analysis

Our data set consists of measuring the time of arrival for electrons or ions created from the reaction between the laser light and atoms. The data typically consist of several runs of 10^4 laser pulses each. We collected data for an equal number of laser shots at fixed laser intensity and polarization direction. A background data set is collected over a similar period with no atomic sample present.
We subtract the background from the signal to remove any residual gas signal and digitizer noise. The time based data is transformed into energy based data using computer analysis program (see Appendix 6.1). In this program temporal data is integrated over energy intervals. We perform consistency checks to look for systematic errors by randomly varying the initial guesses for the parameters. From equation (3.3), the flight time of an electron with kinetic energy $E$ is given by

$$t = \frac{16.9 \text{ (cm)}}{\sqrt{E \text{ (eV)}}} \quad \text{(nsec)}. \quad (3.10)$$

First, we convert the TOF raw data to an energy spectrum. We set the integration bin size to 30 meV and numerically normalized the NIM (Nuclear Instrument Module) pulse amplitude to a one electron count.

$$\text{time} = xinc \cdot (i - 1) - toff, \quad (3.11)$$

$$\text{amp} = \left| \frac{y(i) - \text{baseline}}{\text{NIM pulse amplitude}} \right|, \quad (3.12)$$

where $toff$ is the $t = 0$ or the time of the laser pulse, and $\text{baseline}$ is calculated by taking the average of the last 30 data points. We integrate the energy spectrum using,

$$x(nplot + 1 + i) = ((16.9 \times \text{length})/x(i))^2, \quad (3.13)$$

where $i$ is from 1 to $nplot/2$ and $nplot$ is the number of data points.

We fit the resultant intensity depended data to a power law for the linear portion of a log-log plot, using a nonlinear least squares adjustment (see Appendix 6.2). The linearity is achieved by converting our data to a logarithm using,

$$x(i) = a \log_{10}(x(i)), \quad (3.14)$$
\[ y(i) = a \log_{10}(y(i)). \] (3.15)

We perform a least square fit to a straight line of the form, \( y = a + bx \), and obtain the fit to the slope. The saturation intensity is obtained by measuring the intersect between two lines before and after the turning point, and calculated an \( x \)-axis as a saturation intensity by solving two equations,

\[ y = a_1 + b_1x, \] (3.16)
\[ y = a_2 + b_2x. \] (3.17)

The goodness-of-fit criterion \( \chi^2 \) is

\[ \chi^2 \equiv \Sigma \left( \frac{\Delta y_i}{\sigma_i} \right) = \Sigma \left[ \frac{1}{\sigma_i^2} (y_i - a - bx_i)^2 \right], \] (3.18)

where \( \sigma_i \) is the standard deviation.

The angular distribution measurement follows a similar procedure except the polarization direction is changed for each data set. We measured the distribution at 6° steps, spanning from 0° to 180° range of the polarization angle. The resulting data is fit with a Legendre polynomial of the form,

\[ y = a_0 P_0 + a_1 P_1(x) + \cdots = \sum_{L=0}^{n} a_L P_L(x), \] (3.19)

where \( x = \cos \theta \), with the recursion relationship,

\[ P_L(x) = \frac{1}{L} \left[ (2L - 1)x P_{L-1}(x) - (L - 1)P_{L-2}(x) \right]. \] (3.20)

The deviation of \( \chi^2 \) becomes

\[ \chi^2 \equiv \Sigma \left( \frac{\Delta y_i}{\sigma_i} \right) = \Sigma \left[ \frac{1}{\sigma_i^2} (y_i - y(x_i))^2 \right]. \] (3.21)
We can express the determining equations as a set of \( (n+1) \) equations for the method of least squares which minimize \( \chi^2 \).

\[
\frac{\partial}{\partial a_k} \sum (y_i - y(x_i))^2 = \frac{\partial}{\partial a_k} \sum (y_i - \sum_{j=0}^{n} a_j P_j(x_i) y(x_i))^2 = 0. \tag{3.22}
\]

The solution of these equations yields the values for the coefficients \( a_j \) for which \( \chi^2 \) is a minimum (see Appendix 6.3).
4. NON RESONANT MULTIPHOTON IONIZATION OF ALKALINE-EARTH ATOMS

4.1 Calcium

4.1.1 Electron Energy Spectrum

The electron energy spectrum of calcium with 532 nm excitation at an intensity of $1.7 \times 10^{12} \text{W/cm}^2$ is shown in Fig.13. The laser polarization direction is along the axis of detection. Clearly, the most dominant peak in the electron spectrum is the three-photon ionization of the $4s^2$ ground state of Ca via path (1) (refer to section 2.4.1). Also evident is a peak at an electron energy of 2.11 eV which can be unambiguously assigned to the 6-photon ionization of the $Ca^+$ ion via path (3). The mass spectrum shown in Fig.14 verifies the presence of both singly and doubly ionized calcium at this intensity. Consequently, the results demonstrated that sequential ionization is the predominant mechanism for the production of the observed ion distributions in Ca. This result is consistent with observations made in other alkaline-earth atoms$^{32,34}$. However, this conclusion does not presume that the transition dynamics are restricted to an independent-electron description. Refering to Fig.13(b), we also observe peaks corresponding to the absorption of additional photons producing electrons with energies of 3.21 eV ($h\nu + 0.88 eV$) via path (7) and 4.44 eV ($h\nu + 2.11 eV$) for the $Ca^{2+}(3p^6)$ final state. Moreover, the absorption of additional photons also opens new electron
Fig. 13. The electron spectrum of calcium resulting from 532nm excitation.

The intensity is $1.7 \times 10^{12} W/cm^2$ and the light is polarized along the detector axis. The lower figure is expanded by a factor of 40 in the y-axis.
Fig. 14. The time-of-flight mass spectrum of calcium resulting from (a) 532nm and (b) 1.06μm radiation. The intensities for (a) and (b) are $7.0 \times 10^{11} W/cm^2$ and $7.5 \times 10^{11} W/cm^2$, respectively.
decay channels. Closer examination of the electron spectrum in Fig. 13(b) reveals the presence of a peak at 1.51 eV which is attributable to the Ca$^+$ ion being left in the 3d excited state via path (8). Since one electron is emitted and the other electron is in the excited state, the peak at 1.51 eV immediately shows two electron excitation. Consequently, the MPI of Ca proceeds with some probability for two electron or double excitation. Furthermore, electron peaks at 0.07 and 2.40 eV ($h\nu + 0.07 eV$) are tentatively assigned to the (3+1) photon ionization via path (9) and the (3+2) ionization, respectively. Each of them results in a Ca$^+$ ion being left in a 4p excited state. Our inability to reproduce reliably the relative intensity of the 0.07 eV peak makes this assignment somewhat tentative. We believe that the source of this problem lies in the transmission characteristics of our spectrometer for electrons with energies below 100 meV. However, the observation of the second electron peak in the series with an energy of 2.4 eV ($h\nu + 0.07 eV$) lends some credence to this assignment. It is worth remembering that the measured relative intensities of the peaks are not necessarily representative of the electron-decay branching ratios since our spectrometer views only a small solid angle. This is especially true for the double excitation channels where the angular anisotropy could be quite complex. Future plans include the addition of a 2π electron spectrometer to increase the electron transmission and provide more precise branching ratios.

Fig. 15 shows an electron-energy spectrum of calcium resulting from excitation with 1064 nm photons at an intensity of $3.4 \times 10^{12} W/cm^2$. A number of higher-order peaks are observed in this spectrum but again the dominant peak has an energy of 0.88 eV. This peak is observed to be the major feature at all
Fig. 15. Electron spectrum of calcium resulting from 1.06μm excitation. The intensity is 3.4 \times 10^{12} W/cm^2 and the light is polarized along the detector axis. (b) is expanded by a factor of 16 in the y-axis.
intensities. Unfortunately, at intensities exceeding $10^{11} \text{W/cm}^2$, a problem exists at this wavelength in the absolute interpretation of this peak and its subsequent ATI peaks. This problem is a consequence of the near degeneracy of the electron energies of 0.88 and 0.95 eV, respectively, which are unresolvable with our spectrometer. Obviously the same ambiguity persists for the ATI peaks with electron energies at 2.04 eV ($h\nu + 0.88eV$), 3.21 eV ($2h\nu + 0.88eV$), and 4.36 eV ($3h\nu + 0.88eV$). However, at low intensities, $I < 10^{11} \text{W/cm}^2$, the spectrum is seen to consist of a strong peak at 0.88 eV which is unambiguously assignable to path (2) and its associated ATI peak at 2.04 eV via path (10) (refer to section 2.4.1). At these low intensities the mass spectrum verifies that only singly ionized calcium is present. But as the intensity is raised above $10^{11} \text{W/cm}^2$, the mass spectrum shows that the formation of Ca$^{2+}$ is significant. Likewise, the peaks in the electron spectrum at 0.88 and 2.04 eV broaden and are probably showing the effects due to contributions from the path (4) series. Also, at these intensities the higher ATI peaks at 3.34 eV ($S=2$) and 4.44 eV ($S=3$) start becoming evident. Here $S$ represents the number of additional photons absorbed in the continuum. Presumably, these higher-order processes are a major consequence of path (4) and not of path (2). This would be consistent with higher-order ATI peaks being observed in higher-order ionization processes. Again the main focus of attention in Fig.15(b) is the electron peaks at energies of 0.35, 1.23, 1.51, and 2.68 eV. The assignment of all these peaks characterizes the final state of the ion as excited, that is either a 3d or a 4p state. The 3d series is $\text{Ca}^+ (4s^2) + (6 + S)h\nu \rightarrow \text{Ca}^+ (3d) + e$, where the electron energies at 0.35, 1.51, and 2.68 eV correspond to $S=1,2,$ and 3, respectively. Likewise, the peak
at 1.23 eV corresponds to the ion being left in the 4\textit{p} state following a (6+3) photon absorption with \( S = 3 \). The lowest energy electron from path (14) at 0.07 eV is difficult to observe in the electron spectrum at 1064 nm. The difficulty in detecting this peak may be twofold. First, the transmission problems associated with our spectrometer, as discussed above. Second, with 1064 nm radiation and intensities exceeding \( 10^{11} W/cm^2 \), suppression of the lowest energy peak by the pondermotive potential may become significant. An intensity of \( 3 \times 10^{11} W/cm^2 \) produces a pondermotive potential of 0.03 eV which is becoming comparable to the electron kinetic energy. However, the experimental evidence at 1064 nm, which has a higher order of nonlinearity than at 532 nm, still shows that doubly excited states are playing a role in the multiphoton process. Even though neither excitation scheme results in any detectable amounts of direct ionization, it also does not exclude the possibility that direct ionization could occur under the appropriate circumstances.

4.1.2 Intensity Effects

We experimentally investigated the dependence of the numbers of electrons on the laser intensity. At low intensity, minimum-order perturbation theory predicts that for an \( N \)-photon process the number of counts should scale as \( I^N \). The experimental results for the rapid intensity dependence of the signal for 532 nm and 1064 nm excitation is illustrated in the logarithmic coordinates of Figs.16(a) and 16(b), respectively. The points in Fig.16(a) are measured by integrating the area under the electron peak for the singly ionized 0.88 eV channel [path (1)] and the doubly ionized 2.11 eV channel [path (3)] (refer to section
Fig. 16. Log-log plots of signal versus laser intensity for (a) 532 nm and (b) 1064 nm radiation.
2.4.1). These results were also separately verified by monitoring the total ion signal with a mass spectrometer as a function of intensity at 532 nm. A least-squares fit of the experimental data to a power law for the linear portion of the log-log plot for the 0.88 eV channels yields a slope of $2.9 \pm 0.2$ and a saturation intensity, $I_s$, of $5 \times 10^{10} \text{W/cm}^2$. This result is consistent with the 3-photon ionization of calcium. The dependence beyond the $I_s$ value is a consequence of the expanding focal volume as the intensity is increased. The interaction volume increases as the laser intensity is raised beyond the saturation intensity and consequently more atoms contribute to the ionization signal. Thus the number of electrons slightly increases beyond the saturation intensity. Similarly, the 2.11 eV channel yields a slope of $6.2 \pm 0.4$ and $I_s = 1.2 \times 10^{12} \text{W/cm}^2$, which is in agreement with perturbation scaling laws with 6-photon ionization.

The intensity dependence of the total ion yield for the two charge states for 1064 nm excitation is shown in Fig.16(b). Due to the degeneracy of the electron energies for path (2) and (4) as discussed above, only the 6-photon process could be verified by monitoring the 0.88 eV channel at $I_s < 1.1 \times 10^{11} \text{W/cm}^2$. At these low intensities the ambiguity is removed from the electron spectrum since there is no appreciable yield of the $Ca^{2+}$ ion. The analysis of the total $Ca^+$ ion yield and the 0.88 eV electron channel gives a slope of $5.7 \pm 0.4$ and $I_s = 7.0 \times 10^{10} \text{W/cm}^2$, while analysis of the total $Ca^{2+}$ ion signal yields a slope of $10 \pm 1.5$ and an $I_s = 2.0 \times 10^{11} \text{W/cm}^2$. The slopes here are in good agreement with lowest order perturbation theory and a model consistent with sequential ionization.

Assuming that known scaling laws are valid, the results reported here are in disagreement with the earlier work of Agostini and Petite on the multiphoton
ionization of Ca. Their experiment differs in that the pulse duration of their laser was 50 psec, as opposed to 10 nsec pulses used in this study. Assuming that the saturation intensity is defined as the point at which the atomic population has been reduced to \( e^{-1} \) of its initial value, one obtains a simple expression for the \( N^{th} \) photon process:

\[
I_s = \sigma^{-1/N} \tau^{-1/N},
\]

(4.1)

where \( \sigma \) is the generalized absorption cross section in units of \( cm^{2N} sec^{N-1} \) and \( \tau \) is the pulse duration. Obviously, \( I_s \) increases with decreasing pulse duration and becomes less sensitive to changes in \( \sigma \) and \( \tau \) as the order of nonlinearity becomes large. Using this model to compare the results from excitation with a 10 nsec and 50 psec 1064 nm pulse gives a ratio of \( I_s(50 \text{psec})/I_s(10 \text{nsec}) \) equal to 2.4 and 1.6 for a 6- and 11-photon process, respectively. Table 1 gives the saturation intensity of this work and those estimated from Ref.32 and the ratio of the two results. The measured ratio for the 6-photon ionization of Ca is approximately equal to 10, which differs from the factor of 2.4 predicted by Eq.(4.1). Likewise, the 11-photon ionization of \( Ca^+ \) ion gives an experimental ratio of 4.5 as compared to 1.6 as given by Eq.(4.1). Therefore, the real saturation intensity for shorter pulse width is about 3 - 4 times higher than theoretical value.

The ratio between 3- and 4-photon process for neutral \( Ca \) electron yield is \( C_{a4ph}(S=1)/C_{a3ph}(S=0) = 0.003 \). Likewise, the ratio between 6- and 7-photon process for \( Ca^+ \) ion is measured as \( C_{a7ph}(S=1)/C_{a6ph}(S=0) = 0.043 \). The ratio resulting from these two ratios is \( C_{a4ph}(S=1)/C_{a3ph}(S=0)/C_{a7ph}(S=1)/C_{a6ph}(S=0) = 0.69 \). Therefore, ATI process occurs more easily for higher nonlinearity. For 1064 nm radiation, the electron energies for ionization \( Ca \) and \( Ca^+ \) are 0.88 eV and 0.95 eV, respectively, however we cannot resolve
Table 1. Order of Nonlinearity and Saturation Intensities for Calcium.

<table>
<thead>
<tr>
<th></th>
<th>532-nm</th>
<th>1.06-μm</th>
<th>Ref. 32</th>
<th>$I_5/I_5^*$, 532-nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$N$</td>
<td>$I_s, W/cm^2$</td>
<td>$N$</td>
<td>$I_s, W/cm^2$</td>
</tr>
<tr>
<td>$Ca^+$</td>
<td>$2.9 \pm 0.2$</td>
<td>$5.0 \times 10^{10}$</td>
<td>$5.7 \pm 0.4$</td>
<td>$7 \times 10^{10}$</td>
</tr>
<tr>
<td>$Ca^{2+}$</td>
<td>$6.2 \pm 0.4$</td>
<td>$1.2 \times 10^{12}$</td>
<td>$10 \pm 1.5$</td>
<td>$2 \times 10^{11}$</td>
</tr>
</tbody>
</table>
4.1.3 Angular Distributions

Studies on angular distributions of photoelectrons for multiphoton ionization provide valuable insight into the nature of the atomic transition and useful measurements for comparison with theory. However, as pointed out by Bucksbaum et al.\textsuperscript{71,1}, at these intensities care must be taken in interpreting the origin of the observed angular distributions as purely atomic in nature. The scattering of the atomic electron distributions by the ponderomotive potential can severely alter the detected distribution. This is especially true for low-energy electrons and at long wavelengths since the ponderomotive potential $q^2E_0^2/4m\omega^2$ is proportional to $\omega^{-2}$. Similar scattering effects can be caused by appreciable space charge distributions. For our experiments we kept the atomic density low, and used the laser intensity below the saturation intensity to minimize any such effects and the angular distributions should be only representative of the multiphoton transition.

The electron angular distributions for the singly ionized 0.88 eV channel and the 2.11 eV doubly ionized channel resulting from 532 nm excitation are shown in Fig. 17. The angle in Fig. 17 is measured between the polarization of the light and the direction of the detector. Due to the degeneracy in energy of the different electron channels at high intensity for 1064 nm radiation, it was impossible to derive a meaningful result for any angular distributions except the 0.88 eV channel at low intensity. All the angular distribution studies were performed using linearly polarized light. The electron distributions were recorded by detecting...
Fig. 17. Electron angular distributions for calcium at 532 nm excitation. The open circles are the experimental data points and the solid lines are the results of the fit to the equation of angular distribution. The plots are (a) 0.88 eV channel and (b) 2.11 eV channel.
electrons normal to the laser propagation direction and by rotating the polariza-
tion with respect to the detector axis. The azimuthal angle $\theta$ was varied over $2\pi$
to facilitate the detection of any asymmetries caused by imperfect alignment of
optics. The points in Fig. 17 are the normalized results of the experiments. The
most apparent difference in the shape of the distributions exists between the 0.88
eV channel at 532 nm and 2.11 eV channel at 532 nm. The last process is the
result of a 6-photon absorption while the first is a 3-photon absorption. Conse-
sequently, the 6-photon process, Fig. 17(b), results in angular distributions which
are highly peaked along the polarization direction as compared to the 3-photon
process. This is indicative of the effects produced by solely increasing the order
of nonlinearity in a single electron excitation. In fact, the differences are directly
comparable between the two 0.88 eV channel distributions at the two different
wavelengths. Even comparison of the 2.11 and 0.88 eV channels seems reasonable
considering that both processes originate from $S$ states. The electron angular
distributions for the singly ionized 0.88 eV channel and for the ATI channels
($S=1$ and $S=2$) are shown in Fig. 18. As the degree of nonlinearity increases, the
angular distributions are highly peaked along the polarization direction and the
distribution becomes more and more complicated.

In order to extract some information about the radial elements involved in
these multiphoton angular distributions the experimental data was fitted using
a well-known expression derivable from lowest-order perturbation theory. The-
etically, it can be shown that a nonresonant $N$-photon process results in angular
distributions of the form\textsuperscript{22,76,}

\begin{equation}
I(\theta) \propto \sum_{i=1}^{N} \beta_{2i} P_{2i}(\cos \theta),
\end{equation}
where $P_{2i} (\cos \theta)$ is the Legendre polynomial of order $2i$ and $\beta_{2i}$ are the angular distribution parameters which contain the information on the radial matrix elements. The experimental angular distributions were fit using Eq.(4.2) and the result of the fits are illustrated as solid lines in Figs.17 and 18. Table 2 gives the $\beta_{2i}$ parameters and their standard deviation for each of the three distributions. The parameters are normalized with respect to $\beta_2$ and allowed to fit with the maximum number of $(N+1)$ terms. In order to test the goodness of the fits, analyses were conducted that included extra higher-order parameters in the expression; this resulted in no significant improvement in the fits.

Although the $\beta_{2i}$ angular distribution parameters derived from these fits are not very appealing physically and certainly would benefit from a comparison with theory, some qualitative observations can be made. As described above, as the order of the nonlinearity increases for a nonresonant process the higher-order parameters contribute to the fit resulting in a distribution that is more highly peaked along the laser polarization. This can be seen in Table 2 where the 6-photon processes have a significant contribution from the $\beta_6$ parameter. However as the order of nonlinearity increases the trend in the fits seems to place less emphasis on the contributions from the highest allowable parameters. This is seen in the two, 6-photon fits in Table 2 where both highest-order parameters, $\beta_{10}$ and $\beta_{12}$, are nearly zero within their statistical certainty. This same trend in fits to Eq.(4.2) has been observed by other groups$^{77,78}$ studying nonresonant multiphoton ionization of rare-gas atoms. A simple quantum-mechanical argument would suggest that the transition matrix elements for the final state with the highest orbital angular momentum are zero. Specifically for our experiments final states
Fig. 18. Electron angular distributions for calcium at 1064 nm excitation. The open circles are the experimental data points and the solid lines are the results of the fit to the equation of angular distribution. The plots are (a) 0.88 eV channel, (b) 2.04 eV channel, and (c) 3.21 eV channel.
Table 2. Atomic Parameters from Nonlinear Least-Squares Fit to Equation (2.29).

<table>
<thead>
<tr>
<th>532 nm</th>
<th>Ca + 3hν → Ca⁺ + e(0.88eV)</th>
<th>Ca⁺ + 6hν → Ca²⁺ + e(2.1eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>β₀</td>
<td>0.57(2)</td>
<td>0.56(5)</td>
</tr>
<tr>
<td>β₂</td>
<td>1.00(3)</td>
<td>1.00(9)</td>
</tr>
<tr>
<td>β₄</td>
<td>0.06(3)</td>
<td>0.62(9)</td>
</tr>
<tr>
<td>β₆</td>
<td>0.11(4)</td>
<td>0.44(11)</td>
</tr>
<tr>
<td>β₈</td>
<td>—</td>
<td>0.49(11)</td>
</tr>
<tr>
<td>β₁₀</td>
<td>—</td>
<td>0.00(11)</td>
</tr>
<tr>
<td>β₁₂</td>
<td>—</td>
<td>0.17(12)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>1004 nm</th>
<th>Ca⁺ + 6hν → Ca⁺ + e(0.88eV)</th>
<th>ATI(S=1)</th>
<th>ATI(S=2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>β₀</td>
<td>1.01(9)</td>
<td>0.37(6)</td>
<td>0.36(5)</td>
</tr>
<tr>
<td>β₂</td>
<td>1.00(10)</td>
<td>1.00(9)</td>
<td>1.00(9)</td>
</tr>
<tr>
<td>β₄</td>
<td>2.03(19)</td>
<td>0.62(15)</td>
<td>0.49(13)</td>
</tr>
<tr>
<td>β₆</td>
<td>0.53(13)</td>
<td>0.69(16)</td>
<td>0.55(14)</td>
</tr>
<tr>
<td>β₈</td>
<td>0.24(13)</td>
<td>0.70(17)</td>
<td>0.72(15)</td>
</tr>
<tr>
<td>β₁₀</td>
<td>-0.03(15)</td>
<td>0.32(18)</td>
<td>0.29(16)</td>
</tr>
<tr>
<td>β₁₂</td>
<td>0.14(14)</td>
<td>0.04(20)</td>
<td>-0.01(18)</td>
</tr>
<tr>
<td>β₁₄</td>
<td>—</td>
<td>0.00(17)</td>
<td>-0.12(19)</td>
</tr>
<tr>
<td>β₁₆</td>
<td>—</td>
<td>—</td>
<td>-0.07(17)</td>
</tr>
</tbody>
</table>

† Note: Standard deviations are indicated in parenthesis.
with $L=6$ do not contribute to the transition strength for the 6-photon process. One possible factor that could contribute to this effect extends the arguments used in Ref.1 concerning the centrifugal barrier. However, for our situation the arguments must be modified to consider the effects of the effective potential at a constant electron energy. For circular polarized excitation the effect of the centrifugal barrier for a constant electron-energy channel can result in a suppression of high angular momentum states as the nonlinearity of the process increases.

4.2 MAGNESIUM

4.2.1 Electron Energy Spectrum

The electron energy spectrum of magnesium with 532 nm excitation shown in Fig.19 is at an intensity of $6.0 \times 10^{12} \text{W/cm}^2$. The laser polarization direction is along the axis of detection. The resulting spectrum is composed of lines with energies which are associated with MPI of 4 or more photons. Three series of lines have been well identified: a peak at 1.68 eV, characterizing a 4-photon ionization leaving the ion in the $3s, \, 2S$ state and its ATI series at 4.01 and 6.34 eV; one at 1.91 eV, characterizing the 6-photon ionization to the $Mg^{+}(3p, \, 2P)$ state and its ATI at 4.25 eV; and one at 1.28 eV, characterizing the 7-photon ionization to the doubly ionized $Mg^{2+}(2p^6, \, 1S)$ state. The ATI peak associated with the decay to the $Mg^+(4s)$ state via the $(7+1)$-photon absorption is also well identified at 2.34 eV. But the seven-photon ionization to the $Mg^+(4s)$ state with
Fig. 19. Electron energy spectrum of magnesium at 532 nm radiation. Laser
intensity is $6 \times 10^{12} W/cm^2$. (b) is expanded 4 times.
electron peaks at 0.01 eV was not detected because of the transmission characteristics of our spectrometer at low energies. The most dominant peak in the electron spectrum is at an electron energy of 1.68 eV which is the four-photon ionization of the $3s^2$ ground state of Mg via path (1) (refer to section 2.4.2). The peak at an electron energy of 1.28 eV can be unambiguously assigned to the 7-photon ionization of the $Mg^+$ ion via path (3). The mass spectrum shown in Fig.20 verifies the presence of singly ionized magnesium at this intensity. Also, a strong signal appears at the position of $Mg^{2+}$, but the presence of $Mg^{2+}$ cannot be verified because background carbon ions overlap with doubly ionized magnesium. Fig.21 is an electron spectrum of magnesium and shows that the branching ratio of $Mg^{2+}$ becomes larger as the laser intensity increases. Consequently, the results demonstrate that sequential ionization is the predominant mechanism for the production of the observed ion distributions in magnesium. This result is consistent with our observations in calcium and other alkaline-earth atoms. Again, this conclusion does not presume that the transition dynamics are restricted to an independent-electron description. Referring to Fig.21, we also observe peaks corresponding to the absorption of additional photons producing electrons with energies of 4.01 eV ($h\nu + 0.88 eV$) via path (7). The absorption of two additional photons ($S=2$) opens one more ATI peak at 6.34 eV via path (9). Here $S$ represents the number of additional photons absorbed in the continuum. Moreover, the absorption of two more additional photons also opens new electron decay channels. Closer examination of the electron spectrum in Fig.21 reveals the presence of a peak at 1.91 eV which is attributable to the $Mg^+$ ion being left in the $3p$ excited state via path (8). Consequently, the MPI of Mg proceeds
Fig. 20. Mass spectrum of magnesium at different intensity and wavelength:
(a) $I = 3.9 \times 10^{11}$, 532 nm, (b) $I = 5.7 \times 10^{12}$, 532 nm, and (c) 
$I = 2.2 \times 10^{12}$, 1064 nm.
Fig. 21: Electron energy spectrum of magnesium at 532 nm radiation: (a) $I = 1.1 \times 10^{12} \text{W/cm}^2$, (b) $I = 3.2 \times 10^{12} \text{W/cm}^2$, and (c) $I = 5.4 \times 10^{12} \text{W/cm}^2$. 
with some probability for two electron or double excitation. Furthermore, the electron peak at 2.34 eV \((h\nu + 0.01eV)\) is assigned to the \((4+4)\) photon ionization which results in a \(Mg^+\) ion being left in a 4s excited state. Our inability to produce the 0.01 eV peak makes this assignment somewhat tentative. However, the observation of the second electron peak in the series with an energy of 2.4 eV \((h\nu + 0.07eV)\) lends some credence to this assignment.

Figure 22 shows an electron-energy spectrum of magnesium resulting from excitation with 1064 nm photons. A number of higher-order ATI peaks are observed in this spectrum as the intensity increases and again the dominant feature is the 3s peak with an energy of 0.51 eV. This peak is observed to be the major feature at all intensities. Again the main focus of attention in Fig.22 are the electron peaks at energies of 0.75, 1.17, and 1.91 eV. The assignment of all these peaks characterizes the final state of the ion as excited, that is either a 3p or a 4s state. The 3p series is \(Mg(4s^2) + (7 + S)h\nu \rightarrow Mg^+(3p) + e\), where the electron energies at 0.75, and 1.91 eV correspond to \(S=4\) and \(5\), respectively. Likewise, the peak at 1.17 eV which is ATI of 0.01 eV peak, corresponds to the ion being left in the 4s state following a \((7+S)\) photon absorption with \(S=8\). The lowest energy electron at 0.01 eV is difficult to observe in the electron spectrum at 1064 nm with our spectrometer. Also evident is a peak at an electron energy of 1.28 eV which is assigned to the 14-photon ionization of the \(Mg^+\) 3s ground state to the \(Mg^{2+}\) 2p\(^6\) ground state via path (4). This is the first ATI peak associated with the lowest order 13-photon ionization at 0.11 eV.

Comparing Figs.21 and 22, the peaks for the excited states 3p or 4s of magnesium ion at 1064 nm are not as significant as in the case of 532 nm radiation.
Fig. 22. Electron energy spectrum of magnesium at 1064 nm radiation: (a) $I = 1.6 \times 10^{12} W/cm^2$, (b) $I = 1.0 \times 10^{12} W/cm^2$, and (c) $I = 5.0 \times 10^{11} W/cm^2$. 

$Mg^+ (3s), N = \bar{N}$
Instead, the ATI series of the type 7+1, 7+2, and 7+3 decaying to the 3s ground state of $Mg^+$ ion is more prevalent than with 532 nm excitation. This is understood by considering the number of photons absorbed in the process. At 1064 nm radiation, the MPI processes are

\[
Mg(3s^2) + 7\nu \rightarrow Mg^+(3s) + e(0.51eV)
\]

\[
Mg(3s^2) + 11\nu \rightarrow Mg^+(3p) + e(0.75eV)
\]

\[
Mg(3s^2) + 14\nu \rightarrow Mg^+(4s) + e(0.01eV)
\]

\[
Mg^+(3s) + 14\nu \rightarrow Mg^{2+}(2p^6) + e(0.11eV).
\]

Likewise, the MPI processes at 532 nm radiation are

\[
Mg(3s^2) + 4\nu \rightarrow Mg^+(3s) + e(1.68eV)
\]

\[
Mg(3s^2) + 6\nu \rightarrow Mg^+(3p) + e(1.91eV)
\]

\[
Mg(3s^2) + 7\nu \rightarrow Mg^+(4s) + e(0.01eV)
\]

\[
Mg^+(3s) + 7\nu \rightarrow Mg^{2+}(2p^6) + e(1.28eV).
\]

For the processes at 532 nm radiation, the production of excited states needs two or three more photons than the ground state $Mg^+(3s)$, and the number of counts for excited states are comparable to ATI peaks of $Mg^+(3p)$ for the same nonlinearity. For 1064 nm radiation, the production of a $Mg^+(3s)$ ion needs 7 photons, while $Mg^+(3p)$, $Mg^+(4s)$, and $Mg^{2+}(2p^6)$ needs 11, 14, and 14 photons, respectively. Therefore, the 7+1, 7+2, and 7+3 ATI peaks of $Mg^+(3s)$ state all need a fewer number of photons than any of the processes leading to excited state production. The dominance of the ATI peaks in $Mg$ is a consequence
of the large separation of ground and excited states in $Mg^+$ as compared to $Ca^+$. ATI may also contribute to fast depletion of 3p and 4s states. However, the experimental evidence at 1064 nm, which has a higher order of nonlinearity than at 532 nm, has peaks at 0.75 eV and 1.17 eV which are 3p and 4s excited states, respectively, and shows that doubly excited states are still playing a role in the multiphoton process. Even though neither excitation scheme results in any detectable amounts of direct ionization, it also does not exclude the possibility that direct ionization could occur under the appropriate circumstances.

4.2.2 Intensity Effects

The intensity dependence $I^N$ for an N-photon process for magnesium with 532 nm excitation is illustrated in the log-log plots of Fig.23. The points in Fig.23 are measured for the singly ionized 1.68 eV channel [path (1)] and its ATI [path (7)] (refer to 2.4.2). The ionization of Mg was also separately verified by monitoring the total ion signal with a mass spectrometer as a function of intensity at 532 nm and 1064 nm radiation. For doubly ionized $Mg^{2+}$ ion, the mass spectrum was overlapped with background carbon ions, and could not be analyzed. Analysis of the linear portion of the log-log plot for the 1.68 eV channels gives a slope of $4.2 \pm 0.5$ and a saturation intensity, $I_s$, of $3.2 \times 10^{11} W/cm^2$. This result is consistent with the perturbative scaling for the 4-photon ionization of magnesium. Similarly, the 4.01 eV channel yields a slope of $5.3 \pm 0.7$ and $I_s = 3.9 \times 10^{11} W/cm^2$. $Mg^{2+}$ ions are analyzed via the 1.28 eV channel which have the process of path (3): $Mg^+(3s) + 7\hbar\nu \rightarrow Mg^{2+}(2p^6) + e(1.28eV)$. The saturation intensity of the $Mg^{2+}$ ion ($4.1 \times 10^{11} W/cm^2$) is slightly higher than the saturation intensity
Fig. 23. Log-log plots of signal versus laser intensity for magnesium at 532 nm; 
(a) 1.68 eV channel and (b) 4.01 eV channel.
$I_s = 2.8 \times 10^{11} W/cm^2$ of the total $Mg^+$ ion yield curve. Also, $Mg^{2+}$ ions were not produced at the saturation intensity of $Mg^+$ ion. Therefore, as we discussed in section 2.3, the interaction volume consists of $Mg^+$ ions only, and no neutral Mg atoms exist at the focal point where the atomic beam interacts with the laser. This implies that $Mg^{2+}$ ions are formed from $Mg^+$ ions and verifies a sequential ionization scheme. The intensity dependence of the $Mg^{2+}$ ion production was also measured. The slope $d\log N^{2+}/d\log I = 5.2 \pm 1.5$ is close to the number 7 which is expected for a 7-photon ionization of ions. This also supports the assumption of production of $Mg^{2+}$ ions via sequential ionization. This conclusion is in agreement with previous experiments performed with Ca. The intensity dependence of the total ion yield for singly charged states for 532 nm excitation is shown in Table 3. The analysis of the total $Mg^+$ ion signal yields a slope of $4.4 \pm 0.6$ and an $I_s$ of $2.8 \times 10^{11} W/cm^2$ for the 4-photon ionization. The slope here is also in good agreement with lowest order perturbation theory and a model consistent with sequential ionization. The laser intensity $I = 2.8 \times 10^{11} W/cm^2$ is the saturation intensity at which a marked change appears in the slope. This situation is a typical effect which occurs in MPI experiments when the ionization probability becomes unity, leading to the depletion of atoms in the ionization volume. The intensity dependence beyond the $I_s$ value arises from ions formed in the expanding interaction volume when $I$ is increased.

The intensity dependence of the electron yield of 0.51 eV and a series of ATI at 1064 nm excitation are shown in Fig.24. Figure 24(a) is the log-log plot of 0.51 eV peak. The slope is measured to be $6.7 \pm 1.2$ and consistent with the 7-photon ionization of Mg neutrals. For the 1.68 eV peak which is a 7+1 ATI process,
Table 3. Order of Nonlinearity and Saturation Intensities for Magnesium.

### 532 nm

<table>
<thead>
<tr>
<th>Ionization Process</th>
<th>N</th>
<th>$I_S$ (GW/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Mg(3s^2) + 4\nu \rightarrow Mg^+(3s) + e(1.68)$</td>
<td>4.2(5)</td>
<td>320</td>
</tr>
<tr>
<td>$Mg(3s^2) + (4 + 1)\nu \rightarrow Mg^+(3s) + e(4.01)$</td>
<td>5.3(7)</td>
<td>390</td>
</tr>
<tr>
<td>$Mg^+(3s) + (4 + 3)\nu \rightarrow Mg^{2+}(2p^5) + e(1.28)$</td>
<td>5.2(15)</td>
<td>410</td>
</tr>
<tr>
<td>Total $Mg^+$ Yield</td>
<td>4.4(6)</td>
<td>280</td>
</tr>
<tr>
<td>Total $Mg^{2+}$ Yield</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

### 1064 nm

<table>
<thead>
<tr>
<th>Ionization Process</th>
<th>N</th>
<th>$I_S$ (GW/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Mg(3s^2) + 7\nu \rightarrow Mg^+(3s) + e(0.51ev)$</td>
<td>6.7(12)</td>
<td>430(140)</td>
</tr>
<tr>
<td>$Mg(3s^2) + (7 + 1)\nu \rightarrow Mg^+(3s) + e(1.68ev)$</td>
<td>7.8(9)</td>
<td>400(100)</td>
</tr>
<tr>
<td>$Mg(3s^2) + (7 + 2)\nu \rightarrow Mg^+(3s) + e(2.84)$</td>
<td>9.2(16)</td>
<td>370(100)</td>
</tr>
<tr>
<td>$Mg(3s^2) + (7 + 3)\nu \rightarrow Mg^+(3s) + e(4.01)$</td>
<td>9.7(18)</td>
<td>430(110)</td>
</tr>
<tr>
<td>Total $Mg^+$ Yield</td>
<td>6.9(13)</td>
<td>250</td>
</tr>
<tr>
<td>Total $Mg^{2+}$ Yield</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

† Note: Standard deviations are indicated in parenthesis.
Fig. 24. Log-log plots of signal versus laser intensity for (a) 7-, (b) 8-, (c) 9-, and (d) 10-photon ionization for magnesium at 1064 nm radiation.
the slope is $7.8 \pm 0.9$ and is consistent with the 8-photon ionization process. The $2.84 \text{ eV (7+2 ATI)}$ and $4.00 \text{ eV (7+3 ATI)}$ peaks have slopes of $9.2 \pm 1.6$ and $9.7 \pm 1.8$, respectively, consistent with the values 9 and 10 predicted by lowest order perturbation theory. Peaks associated with the electron decay to the $3p$ and $4s$ excited states of $Mg^+$ appear at $0.75 \text{ eV}$ and $1.17 \text{ eV}$, respectively, in the electron energy spectrum. But, since the peak intensities are so small we are unable to analyze their intensity effect. Note that $I_s$ for 7-photon and ATI series are the same with 1064 nm excitation. This suggests that the 7-photon ionization and its ATI processes occur at the same time in the interaction region.

Table 4 gives the saturation intensity of this work and those calculated from equations (2.11) and (2.13), and the ratio of the two results. For the calculated values, the atomic radius for hydrogen, magnesium, and calcium are $0.5\lambda$, $1.6\lambda$, and $2.0\lambda$, respectively, and the ionization potential are $13.6 \text{ eV}$, $7.6 \text{ eV}$, and $6.1 \text{ eV}$, respectively, in equation (2.11). We also, cited the value for $\lambda^{(H)}$ from Ref.41. The ratio of the saturation intensity between measured and calculated values for $Mg$ with 532 nm and 1064 nm radiation is 1.6 and 0.9, respectively. These numbers are close to 1 and show that the experimental results are in good agreement with theoretical calculations. The same ratio for the $Mg^+$ ions with 532 nm is only 0.1, and means that $Mg^{2+}$ ions are produced by the factor of 10 more than the estimated value. Also, the ratio between 4-photon ionization and its ATI process for $Mg$ neutral is measured as $\frac{M_{g_{4+}}(S=1)}{M_{g_{4+}}(S=0)} = 0.04$. Likewise, the ratio between 7- and 8-photon process for $Mg^+$ ion is measured as $\frac{M_{g_{7+}}(S=1)}{M_{g_{7+}}(S=0)} = 0.15$. This yields a ratio, $\frac{M_{g_{8+}}(S=1)}{M_{g_{4+}}(S=0)} = 0.27$. Again verifying that, ATI processes occur more easily for higher nonlinearity.
Table 4. Comparison of Saturation Intensities for Calcium and Magnesium.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Equation (2.14)</th>
<th>(unit: W/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$I_s(532\text{nm})$</td>
<td>$I_s(1064\text{nm})$</td>
</tr>
<tr>
<td>Ca⁺</td>
<td>$5.0 \times 10^{10}$</td>
<td>$7.0 \times 10^{10}$</td>
</tr>
<tr>
<td>Ca²⁺</td>
<td>$1.2 \times 10^{12}$</td>
<td>$1.3 \times 10^{12}$</td>
</tr>
<tr>
<td>Mg⁺</td>
<td>$3.2 \times 10^{11}$</td>
<td>$2.5 \times 10^{11}$</td>
</tr>
<tr>
<td>Mg²⁺</td>
<td>$4.1 \times 10^{11}$</td>
<td>-</td>
</tr>
<tr>
<td>Sr⁺</td>
<td>$1.0 \times 10^{12}$</td>
<td>-</td>
</tr>
<tr>
<td>Sr²⁺</td>
<td>$5.0 \times 10^{12}$</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>$I_s(1064)/I_s(532)$</th>
<th>$I_s^<em>(1064)/I_s^</em>(532)$</th>
<th>$I_s(532)/I_s(532)$</th>
<th>$I_s(1064)/I_s(1064\text{nm})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca⁺</td>
<td>1.4</td>
<td>5.7</td>
<td>1.2</td>
<td>0.3</td>
</tr>
<tr>
<td>Ca²⁺</td>
<td>1.1</td>
<td>3.9</td>
<td>0.7</td>
<td>0.2</td>
</tr>
<tr>
<td>Mg⁺</td>
<td>0.8</td>
<td>1.4</td>
<td>1.6</td>
<td>0.9</td>
</tr>
<tr>
<td>Mg²⁺</td>
<td>-</td>
<td>2.8</td>
<td>0.1</td>
<td>-</td>
</tr>
<tr>
<td>Sr⁺</td>
<td>-</td>
<td>-</td>
<td>3.7</td>
<td>-</td>
</tr>
<tr>
<td>Sr²⁺</td>
<td>-</td>
<td>-</td>
<td>3.7</td>
<td>-</td>
</tr>
</tbody>
</table>

4.2.3 Angular Distributions

Shown in Figs. 25 and 26 are the angular distributions of Mg\(^+(3s)\) and its ATI peaks with 532 nm excitation. Each of these distributions corresponds to an electron peak with one additional photon absorbed. Note, in Table 5 that higher peaks demand more terms in the expansion because the angular distribution contain terms in the Legendre polynomial of order \(2i\). The \(\beta_{2i}\) coefficients were obtained from the experimental data by using a least-square fitting procedure and are presented with standard deviations in Table 5 for each of the distributions. The coefficient \(\beta_2\) has been normalized to unity. From the \(\beta_{2i}\) coefficients in Table 5, the best fit for ATI becomes more complex as the atom absorbs more photons above the threshold level.

The electron angular distributions for the singly ionized 1.68 eV channel and its ATI series resulting from 532 nm excitation are shown in Fig. 25. The points in Fig. 25 are the normalized results of the experiments. The experimental angular distributions were fit using the equation of angular distribution and the result of the fits are illustrated as solid lines. The most apparent difference in the shape of the distributions is that as the number of photons increases the angular distributions become more peaked along the polarization direction and has more turning points. Fig. 25(a) shows the angular distribution of Mg\(^+(3s)\) which is a 4-photon process. There are maxima at 0°, 90°, and 180° of the azimuthal angle. The angles 0° and 180° are for the laser polarization direction parallel to the detector axis and the angle 90° is the perpendicular direction. Therefore, we see that the ionization probability is maximum when the polarization direction
Table 5. Atomic Parameters from Nonlinear Least-Squares Fit for Magnesium.

<table>
<thead>
<tr>
<th>532 nm</th>
<th>$Mg + 4h\nu \rightarrow Mg^+ + e(1.68\text{ev})$</th>
<th>AT1(S=1)</th>
<th>AT1(S=2)</th>
<th>AT1(S=3)</th>
<th>AT1(S=4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta_0$</td>
<td>1.03(10)</td>
<td>0.41(4)</td>
<td>0.77(16)</td>
<td>1.10(27)</td>
<td>0.93(11)</td>
</tr>
<tr>
<td>$\beta_2$</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
</tr>
<tr>
<td>$\beta_4$</td>
<td>1.20(17)</td>
<td>0.83(12)</td>
<td>0.67(25)</td>
<td>0.64(31)</td>
<td>0.56(16)</td>
</tr>
<tr>
<td>$\beta_6$</td>
<td>-0.26(12)</td>
<td>0.40(11)</td>
<td>1.10(31)</td>
<td>0.37(36)</td>
<td>0.50(20)</td>
</tr>
<tr>
<td>$\beta_8$</td>
<td>0.39(14)</td>
<td>-0.07(11)</td>
<td>0.58(28)</td>
<td>0.05(39)</td>
<td>0.44(19)</td>
</tr>
<tr>
<td>$\gamma_{10}$</td>
<td>-0.26(12)</td>
<td>0.40(11)</td>
<td>1.10(31)</td>
<td>0.37(36)</td>
<td>0.50(20)</td>
</tr>
<tr>
<td>$\beta_{12}$</td>
<td>-0.04(11)</td>
<td>0.02(31)</td>
<td>-0.08(42)</td>
<td>0.16(21)</td>
<td>0.32(23)</td>
</tr>
<tr>
<td>$\beta_{14}$</td>
<td>-0.07(11)</td>
<td>0.05(39)</td>
<td>0.05(39)</td>
<td>0.44(19)</td>
<td>0.05(25)</td>
</tr>
<tr>
<td>$\gamma_{16}$</td>
<td>-0.04(11)</td>
<td>0.02(31)</td>
<td>-0.08(42)</td>
<td>0.16(21)</td>
<td>0.32(23)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>1064 nm</th>
<th>$Mg + 7h\nu \rightarrow Mg^+ + e(0.51\text{ev})$</th>
<th>AT1(S=1)</th>
<th>AT1(S=2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta_0$</td>
<td>-7.36(77)</td>
<td>-5.12(16)</td>
<td>0.80(10)</td>
</tr>
<tr>
<td>$\beta_2$</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
<td>1.00(1)</td>
</tr>
<tr>
<td>$\beta_4$</td>
<td>-4.32(43)</td>
<td>-0.64(39)</td>
<td>0.25(14)</td>
</tr>
<tr>
<td>$\beta_6$</td>
<td>-2.65(28)</td>
<td>-2.02(83)</td>
<td>0.76(19)</td>
</tr>
<tr>
<td>$\beta_8$</td>
<td>-1.07(11)</td>
<td>-5.90(185)</td>
<td>1.42(24)</td>
</tr>
<tr>
<td>$\beta_{10}$</td>
<td>-3.19(12)</td>
<td>3.97(134)</td>
<td>0.68(21)</td>
</tr>
<tr>
<td>$\beta_{12}$</td>
<td>0.97(14)</td>
<td>-0.55(75)</td>
<td>-0.41(23)</td>
</tr>
<tr>
<td>$\beta_{14}$</td>
<td>-0.72(11)</td>
<td>-0.45(81)</td>
<td>0.00(24)</td>
</tr>
<tr>
<td>$\beta_{16}$</td>
<td>-0.38(69)</td>
<td>-0.36(25)</td>
<td>0.00(24)</td>
</tr>
<tr>
<td>$\beta_{18}$</td>
<td>-0.02(24)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Note: Standard deviations are indicated in parenthesis.*
Fig. 25. Electron angular distributions for magnesium at 532 nm excitation. The circles are the experimental data points and the solid lines are the results of the fit to the equation of angular distribution. The plots are (a) 1.68 eV channel, (b) 4.01 eV channel, (c) 6.34 eV channel, (d) 8.67 eV channel, and (e) 11.0 eV channel.
is parallel to the detector, with a secondary maximum for the normal direction. This secondary maximum is consistent with an even-photon absorption in a single electron picture. Fig.25(b) is the angular distribution associated with the ATI peak of $Mg^+(3s)$ which is a (4+1)-photon process. In this case, the slope of the distribution near $0^\circ$ and $180^\circ$ is steeper than that of Fig.25(a). Also, it shows a minimum at $90^\circ$ which means no ionization process occurs in direction normal to the detection axis as expected for odd-photon absorption. Fig.25(c) is the angular distribution of the (4+2)-photon ATI peak of $Mg^+(3s)$. The slope of the distribution near $0^\circ$ and $180^\circ$ is steeper than in the 4- or (4+1)-photon process, and shows more complex structure than Fig.25(a) and (b). Fig.25(d) shows further complexity. In summary, for an N-photon process the angular distribution is highly peaked along the polarization direction and has more complex structure as N increases. Also for an N-photon process, there is a maximum at $90^\circ$ of azimuthal angle when N is even, and minimum when N is odd. This is indicative of the effects produced by solely increasing the order of nonlinearity in a single electron excitation.

The angular distribution for the singly ionized 0.51 eV channel and its ATI series resulting from 1064 nm excitation are shown in Fig.26. The number of photons absorbed ranges from 7 through 9. Fig.26(a) is the angular distribution for 7-photon ionization. The distribution is not as complex as Fig.25(d) which is also a 7-photon process at 532 nm excitation. Fig.26(b) is a (7+1)-photon ionization and shows uniform oscillation through $0^\circ$ to $180^\circ$ of azimuthal angle. It is quite different from any other angular distributions. This may be understood as a 6-photon resonance 7-photon ionization. When we look at the energy level
Fig. 26. Electron angular distributions for magnesium at 1064 nm excitation. The circles are the experimental data points and the solid lines are the results of the fit to the equation of angular distribution. The plots are (a) 0.51 eV channel, (b) 1.68 eV channel, and (c) 2.84 eV channel.
diagram for magnesium (see Fig.6), there is a 3s5d level at about 7.0 eV which is nearly degenerate with 6-photon absorption and has the correct parity. The order of nonlinearity for 7-, 8-, and 9-photon processes in Table 3 consistent with the scaling law and seems to verify that the level does not act like a 6-photon resonance. The 3s5d level probably playing a role in the angular distribution but calculation is necessary to understand the distribution clearly. Fig.26(c) is a (7+2)-photon ionization and shows steeper slope along the polarization direction and has more complex structure than Fig.26(a) and (b). Like the angular distributions at 532 nm, the distributions at 1064 nm radiation are highly peaked along the polarization direction and have complex structure as N increases in N-photon ionization. Also, there is a maximum at 90° when N is even, and minimum when N is odd. These are the common features for both 532 nm and 1064 nm radiation. However, there are big differences between the two excitation schemes. First, the angular distributions at 1064 nm radiation have distinctive maxima and minima along the direction of polarization in spite of the higher nonlinearity. Also, there is some probability of ionization near the perpendicular direction at 1064 nm while it is nearly zero at 532 nm radiation. This may be a manifestation of the ponderomotive potential at this lower frequency and higher intensity. A detailed interpretation of these distributions requires some future theoretical calculations.

4.2.4 Circular Polarization

As a result of the large intensity gradients in the laser focus\textsuperscript{51} ponderomotive scattering appears in the photoelectron angular distribution measurements made
with linearly polarized light. When the ponderomotive forces are less significant, i.e. for lower intensity, shorter wavelength, or higher electron energy, experiments show that electrons are mostly emitted in the direction of the classical driving force along the laser polarization. The use of circularly polarized radiation at 1064 nm for MPI in magnesium dramatically changes the energy distribution of the final state electrons, suppressing all electrons from threshold to 1 eV (Fig.27). In Fig.27 the intensity of the peaks of same energy are compared with linear polarization and circular polarization. The most severely suppressed peak is at 0.51 eV, which has the lowest electron energy of any peaks in the spectrum. This phenomenon is wavelength dependent; for 532 nm radiation the peaks are not suppressed (Fig.28). The disappearance of the low energy electrons can be explained by the centrifugal barrier, or by a reduction of the transition matrix elements for low electron energy states due to the high orbital angular momentum of the final continuum states (Fig.29). A simple model, based on angular momentum absorption and suppression of the overlap with low energy final states by centrifugal repulsion agrees well with these observations. These results indicate that ATI photoelectrons are produced in single step, nonresonant coherent processes directly from the ground state. To consider the suppression of low energy electrons, consider a S ground state of an atom excited to the continuum by a N-photon nonresonant process. Electric dipole selection rules predict:

\[ L = 0, 1, \ldots, N \]

continuum states for linear polarized light, and

\[ L = m_l = N \]
Fig. 27. Electron energy spectrum of magnesium at 1064 nm radiation with the intensity of $2.0 \times 10^{12} W/cm^2$: (a) linear polarization and (b) circular polarization.
Fig. 28. Electron energy spectrum of magnesium at 532 nm radiation with the intensity of $2.0 \times 10^{12} W/cm^2$: (a) linear polarization and (b) circular polarization.
circular states for circular polarized light. Therefore, circular polarized light accesses $L = N$ continuum states. The effective potential for states of definite angular momentum $l$ is

$$V_{\text{eff}}(r) = -\frac{1}{r} + \frac{l(l + 1)}{2r^2} \, \text{(a.u.)},$$

where the first term is the Coulomb potential and the second term is the centrifugal barrier. Now, consider the transition matrix $M$ for the $N$-photon process,

$$M = (eE)^n \sum_{i_1} \cdots \sum_{i_{n-1}} \left[ \frac{\langle \psi_f|d^+|i_{n-1}\rangle \cdots \langle i_1|d^+|\psi_o\rangle}{\delta E_{n-1} \cdots \delta E_1} \right],$$

where $|i_m\rangle$, $|\psi_o\rangle$, and $|\psi_f\rangle$ are intermediate, initial, and final states, respectively; and the detuning $\delta E_i = E_i - E_0 - m\hbar\nu$. In high order nonresonant process the energy denominator is assumed constant over the summand and sum over the numerator alone. The sum over the intermediate states $|i\rangle < \langle i|$ is replaced with the identity matrix $|1\rangle < \langle 1|$. Then, $M$ reduces to a single matrix element,

$$M \propto \frac{(eE)^n}{(\delta E)^{n-1}} \times \langle \psi_f|d^n|\psi_o\rangle,$$

where $e$ and $E$ are electron discharge and electromagnetic field strength, respectively, and $\delta E$ is the average of the resonance energies. The function $r^n\psi_o$, which must overlap the final wave function in the matrix element, has a maximum probability density around $r = na_o$. Near the ionization threshold the angular momentum centrifugal barrier keeps the low energy electrons out at large $r$ and thus, excludes the final state wave function from this region. Therefore, there will be little overlap with the ground state and the transitions are inhibited. At higher energies, corresponding to absorption of more photons and more angular
Fig. 29. Suppression of multiphoton ionization with circularly polarized light.
momentum, the free electron energy rises rapidly and penetrates more closely to the core. Thus, for larger $n$, $r^n\psi_0$ is peaked at larger $r$, and the overlap integral increases and becomes more significant at higher energy. Therefore, MPI process turns on again for large $n$. In summary, the low energy electrons will be greatly suppressed with low photon energy excitation or high nonlinearity.

4.3 COMPARISONS BETWEEN CALCIUM AND MAGNESIUM ATOMS

Let's compare the electron energy spectrum of calcium and magnesium. The ionization threshold of calcium is 6.11 eV above the $4s^2$ ground state and absorption of 3 photons of 532 nm radiation results in a $Ca^+$ ion being left in a $4s$ excited state. The absorption of an additional photon opens 3 decay channels: one ATI of $Ca^+\,(4s)$, and two excited states of $Ca^+(3d)$ and $Ca^+(4p)$. Therefore, the order of nonlinearity for $Ca^+\,(4s)$ ATI peaks is the same as those of excited 3d and 4p states. The ionization threshold of magnesium is 7.64 eV above the $3s^2$ ground state which is higher than that of calcium by 1.53 eV. The minimum number of photons needed to ionize magnesium atom is 4 photons which results in a $Mg^+(3s)$ ion. Magnesium needs two or more photons to reach the excited states: 4+2 photons for 3p state and 4+3 photons for 4s state. Therefore, the ATI process for $Mg^+(3s)$ with $S=1$ has lower nonlinearity and more probability than reaching the 3p or 4d excited state. When we examine the electron energy spectrum for calcium and magnesium ions, the most dominant peaks in any intensity range are the $Ca^+\,(4s)$ and $Mg^+(3s)$ final states. For
the secondary peaks, $Ca^{2+}(3p^6)$ is dominant and doubly excited peaks are comparable to ATI peaks of $Ca^+ (4s)$. On the contrary, the ATI peak of $Mg^+(3s)$ is more dominant than any other peaks for excited states. The reason is that $Mg^+(3s) + 7h\nu \rightarrow Mg^{2+}(2p^6) + e(1.28eV)$ is a 7-photon process and has higher nonlinearity than the $(4+S)$ ATI process of $Mg^+(3s)$ up to $S=3$. The branching ratio of $Mg^+(3s)$ at 1.91 eV increases with the intensity.

For 1064 nm excitation the most dominant peak in the calcium electron spectrum is $Ca^+ (4s)$, as in the excitation of 532 nm, and the secondary peaks are the ATI peaks associated with the ground state $Ca^+ (4s)$, excited states for single ionization $Ca^+ (3d)$ and $Ca^+ (4p)$, and double ionization $Ca^{2+}(3p^6)$. The branching ratio for excited states and doubly ionized calcium are comparable with ATI for $Ca^+ (4s)$. For magnesium, the most dominant peak is $Mg^+(3s)$ as in the case of 532 nm. But at 1064 nm excitation, magnesium has a series of very strong ATI peaks associated with the $Mg^+$, 3s ground state. The branching ratio of ATI peaks for $Mg^+(3s)$ is much larger than those of excited 3p or 4s states. The reason is that $Mg^+$ ion, the 3p and 4s excited states need 7+4 photons and 7+7 photons, respectively. Because of this higher order of nonlinearity, the transition rate for producing the $Mg^+$ ion, 3p and 4s excited states is less than the ATI peaks for $Mg^+$, 3s ground state.

Table 4 shows the saturation intensity for singly and doubly ionized calcium and magnesium atoms. At 532 nm radiation the experimental values of $I_s$ for singly ionized calcium and magnesium are $5.0 \times 10^{10}W/cm^2$ and $3.2 \times 10^{11}W/cm^2$, respectively, and are factor of 1.2 and 1.6 of the calculated values. At 1064 nm, the experimental values of the saturation intensities for singly ionized calcium and
magnesium ions are $7.0 \times 10^{10} W/cm^2$ and $2.5 \times 10^{11} W/cm^2$, and are factor of 0.3 and 0.9 of the calculated values. Therefore, calcium atom is more easily ionized than predicted. Also, atoms are more easily ionized with 1064 nm radiation. For double ionization, the experimental values of the saturation intensities for calcium atoms are $1.2 \times 10^{12} W/cm^2$ and $1.3 \times 10^{12} W/cm^2$ with 532 nm and 1064 nm, respectively. This is lower than the theoretical values by the factor of 0.7 and 0.2, respectively. This trend is even greater for magnesium atoms. At 532 nm radiation, the saturation intensity is measured to be $4.1 \times 10^{11} W/cm^2$ which is 0.1 times lower than the calculated value from equation (2.13), which means that the magnesium atom is 10 times more easily doubly ionized than predicted. In summary, the experimental data for the order of nonlinearity for singly ionized ions with 532 nm radiation are in good agreement with perturbative scaling laws while those for doubly charged ions are not consistent with the scaling laws. The main conclusion from this data is that double ionization of calcium and magnesium atoms is essentially a stepwise or sequential process involving principally the ground state and the first two excited states of the calcium and magnesium ions.

The possibility of a simultaneous ejection of a number of electrons from an atom by a single photon is due to the inner-electron correlations and is therefore a specific multiparticle effect. This multiple MPI has been studied experimentally for Sr and Ba$^{47}$, rare-gas atoms$^{48,49}$, and for several other atoms. The experimental ratio of the double to single electron ionization probability ($N^{2+}/N^+$) at the laser one-electron saturation intensity $I_s$ is shown in Table 6. The ratio for calcium is 0.001 at the saturation intensity of $5.0 \times 10^{10} W/cm^2$. For mag-
Table 6. Comparisons between Calcium and Magnesium at 532 nm Radiation.

<table>
<thead>
<tr>
<th></th>
<th>( \text{N}^2+ / \text{N}^+ )</th>
<th>( I_{\text{N}^2+} (\text{W/cm}^2) )</th>
<th>( I_{\text{N}^+} (\text{W/cm}^2) )</th>
<th>( I_{\text{N}^2+} / I_{\text{N}^+} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca(^+)</td>
<td>0.001</td>
<td>2.0 \times 10^{10}</td>
<td>7.0 \times 10^{11}</td>
<td>35</td>
</tr>
<tr>
<td>Ca(^2+)</td>
<td>1.2 \times 10^{12}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mg(^+)</td>
<td>0.03</td>
<td>2.6 \times 10^{11}</td>
<td>3.2 \times 10^{11}</td>
<td>1.2</td>
</tr>
<tr>
<td>Mg(^2+)</td>
<td>4.1 \times 10^{11}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Xe</td>
<td>1.2 \times 10^{13}</td>
<td>0.015</td>
<td></td>
<td>Ref. 6</td>
</tr>
<tr>
<td>Kr</td>
<td>2.5 \times 10^{13}</td>
<td>0.007</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ar</td>
<td>3.0 \times 10^{13}</td>
<td>0.006</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ne</td>
<td>4.1 \times 10^{13}</td>
<td>0.0015</td>
<td></td>
<td></td>
</tr>
<tr>
<td>He</td>
<td>6.2 \times 10^{14}</td>
<td>0.001</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: \( I_{\text{N}^+} \), \( I_{\text{N}^2+} \) are minimum laser intensities to produce singly and double charged ions, respectively.
nesium, the ratio of $Mg^{2+}$ to $Mg^{+}$ ion yield is 0.03 at the saturation intensity of $3.2 \times 10^{11} \text{W/cm}^2$, which is higher than calcium by a factor of 30. The experimental value of this ratio was found to be $10^{-2} - 10^{-3}$ for inert atoms\textsuperscript{49}. Figure 30 shows the relative ratio of $Ca_{3ph}(S = 0)$, $Ca_{4ph}(S = 1)$, $Ca_{6ph}^+(S = 0)$, and $Ca_{7ph}^+(S = 1)$. Also, shown is the ratio of $Mg_{4ph}(S = 0)$, $Mg_{5ph}(S = 1)$, $Mg_{7ph}^+(S = 0)$, and $Mg_{8ph}^+(S = 1)$. The ratios of the lowest order process and its ATI process for Ca and Mg are $Ca_{4ph} = 0.003$, $Ca_{7ph}^+ = 0.04$, $Mg_{5ph} = 0.04$, and $Mg_{8ph}^+/Mg_{7ph}^+ = 0.15$. By comparison, we can see that magnesium is more easily doubly ionized than calcium and this supports the importance of atomic structure in multiphoton ionization. Another related peculiarity to point out is the laser intensity needed to doubly ionize atoms. The production of the double-charged ions of calcium (6-photon process) can be induced by a laser intensity 35 times higher than the single-electron ionization (3-photon process). In the case of magnesium, the production of doubly charged ions (7-photon process) can be induced by an intensity only 1.2 times larger than that required for single-electron ionization (4-photon process). If we compare the minimum laser intensity needed to produce singly charged ions, magnesium needs more than ten times higher intensity than calcium. But for the doubly ionized ions, the production of $Mg^{2+}$ needs only half of the intensity of $Ca^{2+}$. The reason that $Mg^+$ ion needs higher laser intensity than $Ca^+$ ion is clear because $Mg^+$ is 4-photon process while $Ca^+$ is 3-photon process. But the reason that $Mg^{2+}$ ion needs less intensity than $Ca^{2+}$ ion is not straight forward. The first reason is that the ratio of number of photons necessary for $Mg^{2+}/Mg^+ = \frac{7}{4}$ is less than that of $Ca^{2+}/Ca^+ = \frac{9}{3}$. The second reason is that in the case of $Mg^+$ ion, 3p and 4s excited states open more channels than
Fig. 30. The ionization ratio between (N)- and (N+1)-photon ionization of singly- and doubly-charged ions for both Ca and Mg atoms.
those of Ca, and these channels seem to affect the saturation intensity.

Tables 1 and 3 give the order of nonlinearity and saturation intensities for calcium and magnesium, respectively. For 532 nm radiation, the experimental data for the order of nonlinearity for singly- and doubly-charged calcium ions which are 3- and 6-photon ionization processes are $2.9 \pm 0.2$ and $6.2 \pm 0.4$, respectively. This is in good agreement with perturbative scaling laws. The perturbative scaling law is also valid with 1064 nm radiation. Likewise, it is found to be valid for Mg atoms. A comparison for the saturation intensities for calcium and magnesium can be made using equation (4.1) with $\sigma^{-1/N} = \left( \Lambda_N^{(H)} \frac{R^2}{H} \left( \frac{E_{118}}{E_A} \right) \right)^{-1}$, as we discussed in section 2.3. At 532 nm radiation, calcium is ionized with 3-photons while magnesium is ionized with 4-photons. Using the equation (4.1), the saturation intensity of magnesium is predicted to be 4.9 times higher than calcium, while the experiment shows 6.4 times higher. This is a satisfactory agreement. At 1064 nm radiation, Ca and Mg are ionized with 6- and 7-photons. The ratio of saturation intensity for Mg to Ca is 3.6 and 1.2, respectively, for experiment and calculation. This big discrepancy seems to originate from the higher order of nonlinearity.

Angular distributions for calcium and magnesium are shown in Figs. 17, 18, 25, and 26. The angular distributions of calcium are shown for 3- and 6-photon process at 532 nm, and 6-, 7-, and 8-photon process at 1064 nm radiation. The distributions for magnesium are the 4- through 8-photon processes at 532 nm, and the 7- through 9-photon processes at 1064 nm radiation. By comparing Fig. 17(b) and Fig. 25(c), we see that the angular distributions of the 6-photon process for calcium and magnesium at 532 nm radiation have basically the same
structure: same slope near 0° and 180°, small local maxima at 90°. However, the angular distribution of 7- and 8-photon process at 1064 nm are quite different. Compare Fig. 18(b) and Fig. 26(a) which are 7-photon processes for calcium and magnesium, respectively. Fig. 18(b) has nearly zero probability of ionization and a flat curve near 90° while Fig. 26(a) has a finite probability and turning points in the same region. Fig. 18(c) and Fig. 26(b) which are 8-photon processes for calcium and magnesium show even more differences. Fig. 18(c) has nearly the same distribution as Fig. 18(b) which is a 7-photon process, while Fig. 26(b) shows distinctive oscillations through the azimuthal angle θ. This difference is also seen clearly by comparing Tables 2 and 5. The angular distribution parameter β2i for the 6-photon process at 532 nm are similar between calcium and magnesium, except β0. However, β2i are quite different for 7- and 8-photon processes at 1064 nm. In summary, calcium and magnesium have several common and different features in angular distributions. First, the distribution is highly peaked along the laser polarization as the order of the nonlinearity increases. Also, they have a maximum at 90° of azimuthal angle when N is even, and minimum when N is odd in the N-photon ionization process. However, there are some differences in the angular distributions between calcium and magnesium. If we compare the same number of photons, magnesium has a simpler distribution than calcium. Another difference is that calcium has very weak structure and low probability of MPI in the vicinity of 90° of azimuthal angle, while magnesium has clear structure even when N is 7 or 8. The exact nature of these differences certainly would benefit from a theoretical calculation, but some general observations can be made. The less complex appearance of the magnesium spectrum could reflect
simpler ionization dynamics, that is, more single electron than two electron excitation. This would be consistent with the lower density of doubly excited states at threshold in magnesium as compared to calcium. Also, the higher saturation intensity of magnesium probably shows evidence of ponderomotive scattering for low energy electrons. Assuming a Gaussian field distribution, this will have the effect of smearing the angular distributions.
5. CONCLUSIONS

In this thesis, we presented some recent experimental results on the interaction of intense laser light with alkaline-earth atoms using techniques of multiphoton ionization spectroscopy. Single and double nonresonant ionization of calcium and magnesium atoms have been studied and the electron-energy spectrum with 532 nm and 1064 nm radiation were analyzed at intensities less than $10^{13}\text{W/cm}^2$. We showed conclusively the importance of doubly excited states in the nonresonant excitation with an intense coherent radiation. The role of electron correlations is observed to be significant for the production of singly ionized calcium and magnesium. This is a particular property of alkaline-earth atoms because they have low lying ionic excited states. Single ionization has been shown to result in excited states of the ion and $\text{AII}$ of the ground state of ion. Double ionization was shown to be essentially a sequential process which evolves from the ground state of the ion acting as the initial state for the final process. We would like to stress the importance of understanding the suppression of low energy peaks at low intensity with circular polarized light of alkaline-earth atoms with 1064 nm radiation. Looking into the future, shorter pulse width and more powerful lasers will undoubtedly produce interesting new results. On the theoretical side, no complete multiple ionization theory exists to date. We hope that the experiments, described in this thesis, will help us to develop better theoretical understanding, by providing the insight into the physics of correlated excited states.
REFERENCES


APPENDICES

(A) Electron Energy Analysis Program

0001 PROGRAM ENERGY

C THIS IS THE MAIN PROGRAM FOR THE DATA ANALYSIS OF ELECTRON
C SPECTROMETER. IT ROUTES VARIOUS
C OPTIONS; THIS INCLUDES DATA REDUCTION AND PLOTTING.
C
C ******** THE LINKING INSTRUCTIONS ARE IN ENERGY.COM **********
C
0002 DIMENSION Y(2048), INDEX(50), X(2048)
0003 VIRTUALDATA(4096), BKOND(4096), AMP(4096)
0004 LOGICAL*1 ANS
0005 INTEGER*2 CMD(16)
0006 REAL DELTA, TDIFF, LENGTH
0007 COMMON /HPLOT1/XMAX,XMIN,YMAX,YMIN

C ******** DEFAULT PARAMETERS **********
C
0008 CRIN=30.0 !Energy bin size is 30.0 meV
0009 PAMF=150.0 !Normalization of HPM pulse amplitude
0010 LENGTH=31.50 !Flight tube length is 30.96 cm

C

C 0011 !K=0
C 0012 DATA CMU/'QU','GE','FL','CL','HE','CU','FR','YE','XE',
C 'EN','PA','RA','II','SE','CO','HA'/
C 0013 10 TYPE '+ Option?'
C 0014 ACCEPT 12,1ANS
C 0015 12 FORMAT(A2)
C 0016 B0 20 J=1,16
C 0017 IF(1ANS.EQ.CMD(J)) 0010 30
C 0018 20 CONTINUE
C 0019 30 TYPE '***Error! Not Recognizable Command!!!'
C 0020 do 10 1
C 0021 10 continue

C CHO GE FL CL HE CU FR YE XE IN
C
0022 DATA (9999,200,300,400,500,600,700,900,1100,1300,
C *8500,1400,1500,1600,1700,1800),J
C PA RA II SE CO HA
C
C option UE for setting data
C
C
C 0024 200 call redisk(NSHOTS,X0,XINC,NPTS,NTime,DATAFILE,DATA1,DATA2)
C 0025 201 read=pts

115
DO 202 I=1,INLOT
V(I)=DATASET(I)
X(I)=X0+XINC*(I-1)
202 CONTINUE
DATA=3.
KMIN=NLOT(1)
KMAX=NLOT
IND=1
GOTO 10

C OPTION CL FOR CLEAR SCREEN
C
400 CALL CLEAR
401 CALL CLEAR1
402 CALL SCR1
403 GOTO 10

C OPTION HE FOR SETTING THE MENU
C
500 CALL ENHEL
501 GOTO 10

C OPTION FL FOR PLOTTING SPECTRA
C
300 CONTINUE
302 CALL PLOTS
303 CALL SCR1
304 CONTINUE
305 CALL SCALC(X(IND:),INLOT,KMIN,KMAX)
306 VMAX=VMAX+0.05*(VMAX-VMIN)
307 IF(DATASET.EQ.3) THEN
308 CALL AXIS('TIME, Musc.', 'SIGNAL', 2, 1, 0)
309 IF(DATASET.EQ.4) THEN
310 CALL AXIS('ENERGY, eV', 'COUNTS', 2, 1, 0)
311 CALL PLOT((IND:),X(IND:),INLOT,3,0,1)
312 GOTO 10

C OPTION XE FOR EXPANSION OF THE X-AXIS
C
1100 IRTS=2
1101 CALL CURSOR((IND:),X(IND:),INLOT,INDEX,IRTs)
1102 INDEX=INDEX(2)-INDEX(1)+1
1103 INDEX=INDEX-INDEX(1)-1
1104 XMIN=X(INDEX)
1105 INDEX=INDEX+INDEX(2)-INDEX(1)
1106 INDEX=INDEX-INDEX(1)
1107 INDEX=INDEX+INDEX(2)
1108 INDEX=INDEX-INDEX(1)
1109 INDEX=INDEX+INDEX(2)
1110 CALL CLEAR
1111 GOTO 304

C OPTION CU FOR CURSOR MOVING
C
600 IRTS=50
601 CALL CURSOR((IND:),X(IND:),INLOT,INDEX,IRTs)
602 CALL CLEAR
603 CALL CLEAR1
604 CALL SCR1
605 CALL SCR2
type 355,(1:1:index(l)+index(-1))y(index(1)+index(-1),i=1:1:1pts)
format(' '2(' '12.2(' ')'17.3(2(' ')'12.0,5(' ')'')))
pause
soto 10

C option FR for printing the screen on the printer

700 call cirlsl
call hcopy(1) !Hard copy print
soto 10

C option YE for Expansion of the Y-achs

900 type 1,'What is the new YMIN,YMAX ?'
accept *ymin,ymax
soto 305

C option EN for converting TOF raw data to an energy spectrum

1300 do 1301 i=1:npts
1301 y(i)=data2(i)
1302 if(y(i)=0) goto 1311
1304 data4='Set up data type of energy plot
1305 ibas=nrplot-30
1306 sum=0.0
1308 do 1306 i=ibas,nrplot !Find baseline by taking the
1309 sum=sum+y(i) average of the last 30 points
1310 basin=sum/31.
1311 type 1,'What is t=0 in microseconds ?'
1312 accept *toff
1313 if(toff,en,0.0) toff=en
1314 it=1
1315 do 1320 i=1,nrplot
1319 if(time(t(i)=(i-1))4.0)-toff
1320 if(time(en,0.0) goto 1311
1324 amp(it)=ABS(y(i)-basin)/amp
1325 type 1,'amp=' amp(it)
1326 x(it)=time*1.0e3
1327 it=it+1

1330 continue
1332 do 1330 i=1,nrplot
1334 y(j)=amp(j)
1336 v(nrplot)=0.0
1338 :nrplot=(nrplot-1)*xinc*1.0e3
1340 do 1340 i=1:nrplot
1345 continue
1350 continue

Integrate the energy spectrum
0108  do 1350 i=1,(nplot/2)+1
0109       tmp1=x(nplot+i-1)
0110       x(nplot+i-1)=((16.9*length)/x(i))**2
0111       x(i)=((16.9*length)/tmp1)**2
0112       tmp1=x(nplot+i-1)
0113       y(nplot+i-1)=y(i)
0114       y(i)=tmp1
0115  1350 continue
0116     j=1
0117     delta=chisq/1000.
0118     jmax=nplot-1
0120  5000 continue
0121     if(x(i+1).gt.x(i)+delta*j) goto 5000
0122     y(j)=y(j)+y(i+1)
0123     i=i+1
0125  6100 continue
0126  5100 y(j)=y(j)+(x(1+delta*j-x(i))/(x(i+1)-x(i)))
0127     i=i+1
0128     j=j+1
0129     if(j,j,.lt.jmax) goto 5002
0131  5002 continue
0133     if((j,i).lt.(j+1)+delta*j) goto 6000
0134     do 6100 j=1
0135  6000 y(j)=y(j)+y(i+1)*delta/(x(i+1)-x(i))
0136     j=j+1
0137     if(j,j,.lt.jmax) goto 5002
0139  5002 continue
0141     if((j,i).lt.(j+1)+delta*j) goto 6000
0142  6100 continue
0143  5000 continue
0144     if(j,.eq.1) goto 7000
0145     x(i)=x(i)+delta*(j-1)
0146  7000 continue
0148     do 2000 i=jmax,nplot,1
0149  2000 y(i)=0.0
0150     if(nplot,j,.eq.1) goto 2001
0151  2001 continue
0152     y(i)=y(i)/delta
0153  2001 continue
0154     jmax=0.0
0155     jmax=nplot
0156  300 continue

C
C option BK for subtracting a bkgd from raw data
C
0157  1400 type *,**** Warning! Option only uses RAW DATA. **********
0158     type *, 'This option subtracts background data from the signal.'
0159     type *, 'You must first set the signal data using the GET OPTION.'
0160 call readbk(sh2,n2,inc2,rts2,rt2,dat2,delt2,bkend)
0161     if(nplot.ne.npts2)type *, 'Warning! The # of points are not equal.'
if(nsh2.ne.nshots)type *,'WARNING! The # of shots are not equal,'
if(n0.ne.n02)type *,'ERROR! The # initial are not the same,'
if(ninc1.ne.ninc2)type *,'ERROR! The # of increments are not equal,'

Do you want to normalize the background? (Y/N)'
accept Y/N

if(type.eq.'y'.and.ans.eq.'Y')goto 1402

What is the NORMALIZATION CONSTANT?
accept trynorm
do 1403 j=1,nlot.
bknd(j)=bknd(j)*trynorm
do 1401 i=1,nlot.
data2(i)=data2(i) - bknd(i)
w(i)=data2(i)
continue

BACKGROUND SUBTRACTED,'
soto 10

CONVERTING TO RAW TEMPORAL DATA,'
soto 201

What parameters do you want to change?

What is the new NIM pulse height? (256 max.)'
accept ramp
go to 8500

Current Value=','ramp
What is the new ENERGY BIN SIZE? (in meV)'
accept #chir
go to 8520

Current Value=','chir
What is the new Flight Tube Length? (in cm)'
accept #length
go to 8550

Current Value=','length
Call Plots()
120

0214  goto 305

0215 1700  type * , 'What is the additive correction to x-axis?'
0216   accept * , add2
0217   do 1701 i=1,nplot
0218       x(i)=x(i)+add2
0219 1701  continue
0220  type *, '**** Correction added!!!'
0221  goto 10

0222 1800  type *, 'Printing data!!'
0223   open(unit=6)
0224   nlimit=nplot
0225   do 1801 i=ind,nlimit
0226       write(6,*) i,x(i),w(i)
0227 1801  continue
0228   close(unit=6)
0229  goto 10

0230 9999. call clrscr
0231   call clrscr
0232   call rscrl
0233 9998  format(a1)
0234 9997  FORMAT(15)
0235 9996  format(F10.5)
0236   stop
0237   end
```c
option GE for setting data

type *, 'Option for recalling a stored data set.'
type *, 'Enter the filename!'
accept 210, string, (m(i), i=1, nchar)
format(a, 80=1)
m(nchar+1) = 0
OPEN(UNIT=1, NAME=M, TYPE='OLD', ACCESS='DIRECT',
    ASSOCIATEVARIABLE=NREC, RECORDSIZE=1)
read(1, '1', nets
nrec = 2
do 211 J = 1, nets
    read(i, 'nrec') :: (J)
    nrec = nrec + 1
71 read(i, 'nrec') w(J)
211 continue
close(unit=1)
type *, 'DATA FILE READ'
soto 10

option CL for clear screen

call clrscr
    call clrscr1
    call rscr1
soto 10

option HE for setting the menu

continue
soto 10

option ST for storing x-y array

type *, 'Option for storing x-y array.'
type *, 'Enter the filename!'
accept 610, string, (m(i), i=1, nchar)
format(a, 80=1)
OPEN(UNIT=1, NAME=M, TYPE='NEW', ACCESS='DIRECT',
write(i, 'nrec') :: (J)
write(i, 'nrec') w(J)
```
nrec=nrec+1
continue
close(unit=1)
typetxt,'DATA FILE STORED'
soto 10

option R1 to run the linear fit

700 typetxt,'First line=', last line=''
accept *m, n1
do 710 j=m, n1
k=j+1
x1(k)=x(j)
y1(k)=y(j)
710 continue
nlin1=n1+m+1
c
call linfit(x1, y1, sigma1, nlin1, mode, a, sigma1, b1, s1, b1, r)
c
typetxt,' Y = a + b * X '
typetxt,' a=', a1
typetxt,' b=', b1
typetxt,' sigma1=', sigma1
typetxt,' s1=', s1
soto 10

c
option R2 to run the linear fit

800 typetxt,'First line=', last line=''
accept *m2, n2
do 810 J=m2, n2
k=j+m2+1
x2(k)=x(j)
y2(k)=y(j)
810 continue
nlin2=n2+m2+1
c
call linfit(x2, y2, sigma2, nlin2, mode, a2, sigma2, b2, s2, b2, r)
c
typetxt,' Y = a + b * X '
typetxt,' a=', a2
typetxt,' b=', b2
typetxt,' sigma2=', sigma2
typetxt,' s2=', s2
soto 10

c
c
option FL to plot the data
300  type *, 'Option for plotting data'
            type *, 'TYPE11 for raw data plot'
            type *, 'TYPE12 for combined plot with line 1'
            type *, 'TYPE13 for combined plot with lines 1 and 2'
            accept *, Jflag
            type *, 'xmin'?: xmin= ? ymin= ? ymax= ?
            accept *, 'xminmax'?: xminmax= ?
            call plots()
            call scroll
            call axis('Log10 (Intensity)', 'Log10 (Number of Counts)' , 1, 1, 0)
            call plot(xs, nums, 3, 1)
            if(Jflag .eq. 2) goto 340
            do 320 i = 1, nlin1
                wv1(i) = a1 * b1 * x(i)
                call plot(x, wv1, mlin1, 3, 0, 1)
            if(Jflag .eq. 2) goto 340
            do 350 i = 1, nlin2
                wv2(i) = a2 * b2 * x(i)
                call plot(x, wv2, mlin2, 3, 0, 1)
            goto 340

340  goto 10

900  ysa = -(a2 - a1) / (b2 - b1)
     ysa = a1 * b2 - b1 * a2) / (b2 - b1)
     ysa = 10 ** ysa
     ysa = 10 ** ysa
     type *, 'YSA=' : ysa
            type *, 'YSN=' : ysn
            goto 10

1000 type *, 'This option allows editing of specific data pts'
             type *, 'What is the value of the x in the x-y pair'
             accept *,
             x(n) = 10 ** x(n)
             y(n) = 10 ** y(n)
             type *, '0ld := ', x(n), '0ld w=' , y(n)
type *'What are NEW X, Y?'
accept *'X'(n), 'Y'(n)
type *'New x=' 'X'(n), 'New y=' 'Y'(n)
x(n) = sin10X(n(n))
y(n) = cos10Y(n(n))
auto 10

option GU to stop the program

call clrdr

call rscr

call rscr

stop

end
SUBROUTINE LEGFIT

Purpose

Make a least-squares fit to data with a Legendre Polynomial

\[ Y = A(1) + A(2)X + A(3)(3X^2-1)/2 + \ldots \]

\[ = A(1) + (1, \frac{1}{3} B(2) + \frac{1}{5} B(3)(3X^2-1)/2 + \ldots ) \]

where \( X = \cos(\theta) \)

Usage

Call Legfit(theta,y,sigma,nts,norder,neven,mode,ftest,yfit,sigmaa,simab,chi2)

Description of parameters

\( \theta \) - array of angles (in degrees) of the data points
\( Y \) - array of data points for dependent variable
\( \text{Sigma} \) - array of standard deviations for \( Y \) data points
\( \text{Nts} \) - number of pairs of data points
\( \text{Norder} \) - highest order of polynomial
\( \text{Neven} \) - determines odd or even character of polynomial

- 0 fits only to even terms
- 1 fits only to odd terms (plus constant term)

\( \text{Mode} \) - determines mode of weighting least-squares fit

- 1 (instrumental) weight(i) = 1/sigma(i)*2
- 0 (no weighting) weight(i) = 1
- -1 (statistical) weight(i) = 1/sigma(i)

\( \text{Ftest} \) - array of values of \( f(L) \) for an f test
\( \text{Yfit} \) - array of calculated values of \( Y \)
\( a \) - array of coefficients of polynomial
\( \text{Sigmaa} \) - array of standard deviations for coefficients
\( b \) - array of normalized relative coefficients
\( \text{Simab} \) - array of standard deviations for relative coeff
\( \text{Chisqr} \) - reduced chi square for fit

Subroutines required

Matinv(array,nts,norder,de)

Comments

Dimension statement valid for nts up to 100 and norder 9

Subroutine Legfit(theta,y,sigma,nts,norder,neven,mode,ftest,yfit,sigmaa,simab,chi2)

double precision cosine,beta,alha,simab,chi2

virtual a(1),sigma(1),b(1),simab(1)
virtual weight(100),p(100,20),beta(20),alha(20,20)

Accumulate weights and Legendre polynomials

11

integral
ncoeff=1
Jmax=norder+1
20 do 40 i=1,nets
21 if(mode) 22,27,29
22 if(w(i)) 25,27,23
23 weight(i)=1./w(i)
soto 31
25 weight(i)=1./(-w(i))
soto 31
27 weight(i)=1,
soto 31
29 weight(i)=1./signay(i)**2
31 cosine=cos(0.01745329252*theta(i)):
p(i,1)=1.
p(i,2)=cosine
do 36 l=2,norder
fl=1.
36 p(i,1)=((2.*fl-1.)*cosine*p(i,1)-(fl-1.+1.*u(i-1)))/fl
40 continue

c Accumulate matrices alpha and beta
c
51 do 54 J=1/terms
beta(J)=0.
do 54 k=1/terms
54 alpha(J,k)=0.
61 do 66 i=1/terms
66 J=1/terms
beta(J)=beta(J)+p(i,J)*w(i)*weight(i)
do 66 k=1/terms
alpha(J,k)=alpha(J,k)+p(i,J)*p(i,k)*weight(i)
66 alpha(k,J)=alpha(J,k)
c Delete fine coefficients
c
70 if(neven) 71,91,81
71 do 76 J=3/terms,2
beta(J)=0.
do 75 k=1/terms
alpha(J,k)=0.
75 alpha(k,J)=0.
76 alpha(J,J)=1.
soto 91
81 do 86 J=2/terms,2
81 beta(J)=0.
do 85 k=1/terms
alpha(J,k)=0.
85 alpha(k,J)=0.
86 alpha(J,J)=1.
c
Invert curvature matrix alpha
c
91 do 95 J=1,Jmax
c Calculate coefficients, fit, and chi square

do 115 J=1,nterms
  do 113 k=1,nterms
    a(J)=a(J)+beta(k)*alpha(J,k)
  do 115 i=1,nts
    wfit(i)=wfit(i)+a(J)*f(i,J)
  chi=s=0.
  do 123 i=1,nts
    chi=chi+(y(i)-wfit(i))^2*weight(i)
    free=nts-ncoeff
    chi=chi/chi
  c Test for end of fit
  if(nterms-Jmax)132,151
  if(ncoeff=2)133,134,141
  if(neven)137,137,135
  if(neven)135,137,135
  nterms=nterms+2
  goto 138
  nterms=nterms+1
  ncoeff=ncoeff-1
  chiold=chi
  goto 51
  fvalue=(chi1-chi0)/chi
  if(fvalue(nterms-fvalue)134,143,143
  if(neven)144,146,144
  nterms=nterms-2
  goto 147
  nterms=nterms-1
  ncoeff=ncoeff-1
  Jmax=nterms
  goto 51
  c Calculate remainder of output
  if(mode)152,154,152
  varnce=1.
soto 155
154  varnce=chisor
155  do 156 J=1,INTERMS
156  sigmax(J)=dsort(varnce*alpha(J,J))
157  if(a(1))162,170,163
158  do 166 J=2,INTERMS
159  if(a(J))164,166,164
160  b(J)=a(J)/n(1)
161  sigmax(J)=b(J)*dsort((sigmax(J)/a(J))**2*(sigmax(1)/a(1))**2
162  1-2.*varnce*alpha(J,1)/(a(J)*a(1)))
163  continue
164  b(1)=1.
165  return
166  end
VITA

Dal-Woo Kim
Address: 414 Lake Pointe Drive
Middle Island, New York 11953
Phone: (516)282-4339 office
(516)345-5264 home

Teaching Experience:

1/85 to 5/87 Louisiana State University, Baton Rouge, La.
Teaching Assistant;
Conducted three hour labs twice weekly.
Presented lecture on basic principle of physics.
Directed laboratory experiments.

Research Experience:

1/88 to present Brookhaven National Laboratory, Upton, N.Y.
Junior Research Associate;
Studied the multiphoton ionization of alkaline-earth.

6/87 to 12/87 Louisiana State University, Baton Rouge, La.
Research Assistant;
Developed time-of-flight spectrometer.

1/78 to 12/84 Pohang Iron and Steel Co., Pohang, Korea
Research Staff;
Researched surface flaw detection with He-Ne laser.

Publications:

L. F. DiMauro, Dalwoo Kim, M. W. Courtney, and M. Anselment,

130
Dalwoo Kim and L. F. DiMauro, Proceedings of the Fourth International Symposium on Resonance Ionization Spectroscopy and its Applications

**Education:**

<table>
<thead>
<tr>
<th>Degree</th>
<th>Year</th>
<th>Institution</th>
<th>Notes</th>
</tr>
</thead>
</table>
| Ph.D. | 1989 | Louisiana State University, Baton Rouge | Louisiana State University, Baton Rouge 
Dissertation: *Nonresonant Multiphoton Ionization of Alkaline-Earth Atoms in Intense Laser Fields* |
| M.S. | 1987 | Louisiana State University, Baton Rouge | |
| B.S. | 1973 | Seoul National University, Seoul, Korea | |

**Personal:**

- Born: September 16, 1949
- Birthplace: Pusan, Korea
- Married, One child

**References:**

- Dr. Louis F. DiMauro, Department of Chemistry, Brookhaven National Laboratory, Upton, New York, 11973. Telephone: (516) 282-4323
- Dr. A.R.P. Rau, Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803. Telephone: (504) 388-6841
- Dr. Michael G. White, Department of Chemistry, Brookhaven National Laboratory, Upton, New York, 11973. Telephone: (516) 282-4345
DOCTORAL EXAMINATION AND DISSERTATION REPORT

Candidate: Dalwoo Kim

Major Field: Physics

Title of Dissertation: Nonresonant Multiphoton Ionization of Alkaline-Earth Atoms in Intense Laser Fields

Approved:

[Signatures]

Major Professor and Chairman

Dean of the Graduate School

EXAMINING COMMITTEE:

[Signatures]

Date of Examination: April 19, 1989