ELSEVIER

Journal of Quantitative Spectroscopy & Radiative Transfer 109 (2008) 107–118

Journal of Quantitative Spectroscopy & Radiative Transfer

www.elsevier.com/locate/jqsrt

Two-, three-, and four-photon ionization of Mg in the circularly and linearly polarized laser fields: Comparative study using the Hartree–Fock and model potentials

Gabriela Buica[1], Takashi Nakajima*

Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan

Received 22 January 2007; received in revised form 8 May 2007; accepted 15 May 2007

Abstract

We theoretically study multiphoton ionization of Mg in the circularly polarized (CP) as well as the linearly polarized (LP) laser fields. Specifically two-, three-, and four-photon ionization cross sections from the ground and first excited states are calculated as a function of photon energy. Calculations are performed using the frozen-core Hartree–Fock (FCHF) and also the model potential (MP) approaches and the results are compared. We find that the MP approach provides results as good as or even slightly better than those by the FCHF. We also report the relative ratios of the ionization cross sections by the CP and LP laser fields as a function of photon energy, which exhibit clear effects of electron correlations.

Keywords: Magnesium; Multiphoton ionization; Cross section; Hartree–Fock; Model potential

1. Introduction

The interest for studying multiphoton ionization processes by using a circularly polarized (CP) field increased in the beginning of 1970s because of their potential of providing larger ionization cross sections in comparison to a linearly polarized (LP) field. Indeed, for a single-valence-electron atoms such as H and Cs, it has been shown in Refs. [1,2] that the two- and three-photon ionization cross sections by the CP field are larger than those by the LP field. Soon after that, however, it was realized that this is not always true, especially for multiphoton resonant and near-resonant ionization processes in which more than a few photons are involved [3]. Depending on the atoms and the photon energy, cross sections by the LP field can be larger or smaller than those by the CP field. Reiss gave the correct interpretation of the CP and LP cross section ratio [3]: The ratio could be larger than 1 only for processes involving up to four- or five-photon ionization. This can be understood as a consequence of the following two reasons: First, if the number of photons involved for ionization is more than a few, there is more chance for the LP field than the CP field to be close to resonance

*Corresponding author.
E-mail addresses: gzloh@yahoo.com (G. Buica), t-nakajima@iae.kyoto-u.ac.jp (T. Nakajima).
[1]Permanent address: Institute for Space Sciences, PO Box MG 23, RO 77125 Bucuresti-Magurele, Romania.

0022-4073/$ - see front matter
doi:10.1016/j.jqsrt.2007.05.004

with some bound states during multiphoton absorption. Second, since there are more ionization channels, in terms of the number of the accessible partial waves, for the LP field than the CP field, the sum of all these partial wave contributions could simply become larger for the LP field than the CP field. We note that most of the theoretical studies performed in the above context in those days is mainly for the single-valence-electron atoms and there are no published results for more complex atoms in a CP field.

Time has passed since then, and because of the significant development of theoretical methods and computer powers to calculate atomic structures, it is now possible to calculate multiphoton ionization cross sections for more complex atoms with reliable accuracy. Although there are quite a few theoretical reports for the multiphoton ionization cross sections beyond the single-valence-electron atoms such as Xe, He, Be, Mg, and Ca [4–13], all of them assume the LP field. Although theoretical data for the multiphoton ionization cross sections for complex atoms by the CP field would provide complementary information to those by the LP field for the purpose of understanding the multiphoton dynamics, such data are still missing in the literature.

The purpose of this paper is to present theoretical results for the multiphoton ionization cross sections of Mg by the CP laser field by using the frozen-core Hartree–Fock (FCHF) and model potential (MP) approaches. By comparing the calculated state energies and oscillator strengths, we find that the MP approach is as good as the FCHF approach for the oscillator strengths. As for the state energies, the MP approach gives even better numbers than the FCHF approach. As a result, for the purpose of calculating multiphoton ionization cross sections of Mg, the MP approach could give even slightly better numbers than the FCHF approach. As additional data, we also provide the *ratios* of the multiphoton ionization cross sections by the LP and CP fields, which would be very useful from the experimental point of view, since the *absolute* measurement of the multiphoton ionization cross sections is usually very difficult. The ratio of the multiphoton ionization cross sections of Mg by the CP and LP fields we have obtained exhibits a similar behavior with those reported for the single-valence-electron atoms as well as rare gas atoms [1,2,14,15].

2. Theoretical approach

The Mg atom is a *two-valence-electron* atom; it consists of a closed core (the nucleus and the 10 inner-shell electrons) and two-valence electrons. As it is already mentioned in the literature [16] there are several approaches to solve the Schrödinger equation for one- and two-valence-electron atom in a laser field. Since the general computational procedure has already been presented in Refs. [6,9,10] and the specific details about the atomic structure calculation of Mg have been reported in recent works [11,12], we only briefly describe the method we employ. The field-free one-electron Hamiltonian of Mg^+, $H_a(r)$, is expressed, in a.u., as

$$H_a(r) = -\frac{1}{2}\frac{d^2}{dr^2} - \frac{Z}{r} + \frac{l(l+1)}{2r^2} + V_{\text{eff}}(r), \tag{1}$$

where $V_{\text{eff}}(r)$ is the effective potential acting on the valence electron of Mg^+. r represents the position vector of the valence electron, Z the core charge (= 2 in our case), and l the angular quantum number. Depending on how we describe $V_{\text{eff}}(r)$, we consider two approaches in this paper, the FCHF and the MP methods.

2.1. One-electron orbitals: frozen-core Hartree–Fock approach

The most widely used approach to describe the ionic core would be the FCHF approach. In the FCHF approach the ionic core of Mg^{2+} ($1s^2 2s^2 2p^6$) is given by

$$V_{\text{eff}}(r) \to V_l^{\text{HF}}(r) + V_l^{\text{p}}(r), \tag{2}$$

where V_l^{HF} represents the FCHF potential (FCHFP) and V_l^{p} is the core-polarization potential which effectively accounts for the interaction between the closed core and the valence electrons [6]. Specifically we employ the following form for the core-polarization. $V_l^{\text{p}}(r) = -\frac{\alpha_s}{2r^4}[1 - \exp^{-(r/r_l)^6}]$, in which α_s is the static dipole polarizability of Mg^{2+} and r_l the cut-off radii for the different orbital angular momenta, $l = 0, 1, 2, \ldots$. For the expansion of one-electron orbitals, we employ a B-spline basis set. Thus, solving the Schrödinger equation for the nonrelativistic Hamiltonian defined in Eq. (1) with the FCHFP is now equivalent to the eigenvalue problem.