Coherent phase control in ionization of magnesium by a bichromatic laser field of frequencies \( \omega \) and \( 2\omega \)

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Abstract. We have investigated the coherent phase control on the \( 3p^2 \) autoionizing state (AIS) resonantly coupled with the ground state for Mg through a two- and a four-photon transition simultaneously, using a bichromatic linearly polarized laser field. The frequency is chosen such that the lasers are tunable around resonance with the transition \( 3s^2(^1S^0) \rightarrow 3p^2(^1S^0) \), which implies \( \omega_1 = 2.11 \text{ eV} \) and \( \omega_2 = 4.22 \text{ eV} \). We are interested in the modification of autoionizing (AI) line shape through the relative phase and laser intensities. A strong phase dependence on the total ionization yield and ionization rate is found. We also performed a time-dependent calculation which takes into consideration all the resonant states of the process.

PACS. 32.80.Qk Coherent control of atomic interactions with photons – 32.80.Rm Multiphoton ionization and excitation to highly excited states (e.g., Rydberg states) – 32.80.Dz Autoionization

1 Introduction

This paper treats a particular aspect of the control of photoabsorption, and in particular, ionization through the relative phase of two electromagnetic fields acting simultaneously on an atomic system. The aspect in question is the possibility of altering the line shape of an autoionizing resonance through such phase control. The general features of phase control in photoabsorption have been discussed in the literature extensively [1–7]. The possibility of altering an autoionizing line shape has also been discussed [8] to some extent in theoretical work where it has been shown that one particularly intriguing feature, depending on parameters, is the possibility of turning off the transition to the “discrete” or the continuum part of the resonance [9,10]. To the best of our knowledge, experimental observation of such an effect has not been recorded, one of the reasons perhaps being the unavailability of radiation sources of intensity and frequency suitable for convenient atomic species and resonances.

The case studied in this paper aims at presenting a quantitative analysis of a situation possibly convenient for existing laser sources whose potential in phase control has been tested experimentally. We have thus undertaken the study of the line shape of the \( 3p^2(^1S^0) \) autoionizing resonance of atomic magnesium under the simultaneous excitation by four- and two-photon transitions whose relative phase is varied. This case [11] is somewhat different from, and theoretically a bit more demanding, than the more standard scheme of one- and three-photon transitions, which in any case would not be applicable here owing to the parity of the resonance which is the same as that of the ground state. In addition, we have found that bound states in near resonance with one- or three-photon transitions introduce distortions in the wings of the autoionizing resonance; an effect not a priori obvious but which has to be taken into account for a realistic assessment of the observability of the desired feature. Needles to say that a realistic atomic structure and transitions calculation has been necessary, whose details are discussed in the sections that follow.

Lyras and Bachau [8] studied the phase control in two- and four-photon ionization of the magnesium atom in a bichromatic field of frequencies \( \omega \) and \( 3\omega \), in the vicinity of an autoionizing resonance \(^1D^o\) lying above the first two ionization thresholds. Moreover, they accomplished a full perturbative, time-independent phase control calculation in Mg by interfering three- and one-photon transitions to a single continuum channel \(^3P^o\). They studied the dependence of the ionization rate as a function of the relative phase for different laser intensities, and noticed that the presence of an intermediate resonance may enhance the overall ionization signal.

Kylstra et al. [12] have performed a non-perturbative ab initio one- and two-color calculation of the multiphoton ionization of magnesium, where the laser frequencies are chosen such that the initial state of the atom is resonantly

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coupled to the autoionizing 3p^2, 3s3d, and 4s5s resonances of the atom. Using R-matrix Floquet theory, they studied single photon ionization from the ground state 3s^2 and the 3s3p excited state of magnesium in the vicinity of the 3p^2 AI resonance at non-perturbative laser intensities. A second low-frequency laser couples the 3p^2 and 3s3d AI states. This resonant system was studied experimentally and theoretically in the perturbative domain. Kylstra et al. also investigated the harmonic intensity regime where the process is no longer perturbative.

In Section 2 we present the basic framework related to the study of the phase control in the vicinity of an AI state. In Section 3 we briefly give the ab initio theoretical approach for the description of the magnesium atomic structure. Further details can be found in [13–15]. Finally, in Section 4 we present our results.

2 Basic equations

We study the multiphoton ionization of Mg using a realistic atomic model which describes the time-evolution of the system, investigating the phase coherent effect on the 3p^2 (1S^o) autoionizing state resonantly coupled to the ground state of Mg, simultaneously through two- and four-photon transitions, in the weak field regime. The laser frequency is such that the ground state of the atom is near resonance with the 3s3p (1P^o) (E_{3s3p} ≈ 4.34 eV) state of the Mg atom due to its second harmonic, and with 3s4p (1P^o) (E_{3s4p} ≈ 6.11 eV) due to three-photon absorption from the fundamental.

We consider the magnesium atom consisting of a ground state |g⟩ = |3s^2 1S^o⟩, an autoionizing state |a⟩ = |3p^2 1S^o⟩, and one continuum corresponding to three different values of angular momentum |c_1⟩ = |3s3s 1S^o⟩, |c_2⟩ = |3s3d 1D^o⟩, and |c_3⟩ = |3s2g 1G^o⟩. The AIS is modeled as a discrete state embedded into the continuum and coupled to the 1S^o continuum through the configuration interaction.

In Figure 1 we show the energy diagram of the atomic system interacting with the linearly polarized bichromatic field:

\[ \mathbf{E}(t) = \mathbf{E}_f(t) \exp(i\omega_f t) + \mathbf{E}_h(t) \exp(i\omega_h t + i\varphi) + c.c., \]

consisting of a superposition between the fundamental with frequency \( \omega_f \) and its second harmonic with frequency \( \omega_h = 2\omega_f \). The amplitudes of the two components of the electric field are \( \mathbf{E}_f(t) \), \( \mathbf{E}_h(t) \), and \( \varphi \) is the relative phase between them. The continuum state of energy \( E_0 + 4\omega_f \) can be reached by the two interfering paths shown in Figure 1, namely, the four photon absorption from the fundamental electromagnetic field (Fig. 1, path (i)), and the two-photon absorption of its second harmonic (Fig. 1, path (ii)).

There are a few other processes which might affect the interference process, but for the intensities we consider in this paper they are not significant, therefore we do not need to include them in the calculation. For instance, a three-photon transition with two photons of the fundamental and one of the harmonic could lead to the same final continuum energy. These processes, however, could only influence the background signal, and not the interference process, since the final continuum state belongs either to a \( 1P^o \) or to a \( 1F^o \) state. We have estimated the transition amplitudes for these three-photon transitions and for values of the laser intensities involved in our calculation, and we found out that the contribution to the ionization signal is about two orders of magnitude smaller than the contribution due to the processes presented in Figure 1. One can therefore conclude that these three-photon processes do not affect the process studied here and we neglect them. The dominant processes are decided by the value of the frequency and intensities involved in the calculation. For the same reason, Raman-type processes are ignored due to the fact that they represent higher-order processes with respect to the electric field. We have used atomic units throughout this work. The atomic unit used for the intensity of laser field is \( I_0 = 14.037 \times 10^{16} \text{ W/cm}^2 \).

A four photon transition from the \( |3s^2 1S^o⟩ \) state into the continuum leads to a final state containing \( |3s3s 1S^o⟩ \), \( |3s3d 1D^o⟩ \), and \( |3s2g 1G^o⟩ \) partial waves, while a two-photon transition into the continuum leads only to \( |3s3s 1S^o⟩ \) and \( |3s3d 1D^o⟩ \) partial waves. Thus, only the transition amplitude to the \( 1S^o \) and \( 1D^o \) continua is modulated through the interference with the two-photon amplitude. The transition amplitude to the \( 1G^o \) continuum remains unaffected by the quantum interference, and therefore only contributes to the background of the total ionization signal.

We consider the Schrödinger equation:

\[ i\frac{\partial \Psi(t)}{\partial t} = [H_a + D(t)]\Psi(t), \]

where \( H_a \) represents the atomic Hamiltonian. The atomic system is treated as a two active electron system: the atomic core (the nucleus and the 10 inner-shell electrons), and two valence electrons. More details about the atomic structure calculation are given in the next section. Within the semiclassical formalism, the interaction between the atom and the bichromatic field is described in the length gauge, and in the dipole approximation by