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The multiphoton ionization rate and the energy shift of atoms interacting with weak dichromatic fields with commensurate frequencies are simple functions of the phase difference

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Abstract. By implementing a time-independent, nonperturbative many-electron, many-photon theory (MEMPT), cycle-averaged complex eigenvalues were obtained for the He atom, whose real part gives the field-induced energy shift, $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$, and the imaginary part is the multiphoton ionization rate, $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi)$, where ω is the frequency, F is the field strength and φ is the phase difference. Through analysis and computation we show that, provided the intensities are weak, the dependence of $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi)$ on φ is simple. Specifically, for odd harmonics, Γ varies linearly with $\cos(\varphi)$ whilst for even harmonics it varies linearly with $\cos(2\varphi)$. In addition, this dependence on φ holds for $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$ as well. These relations may turn out to be applicable to other atomic systems as well, and to provide a definition of the weak field regime in the dichromatic case. When the combination of (ω_1, F_1) and (ω_2, F_2) is such that higher powers of $\cos(\varphi)$ and $\cos(2\varphi)$ become important, these rules break down and we reach the strong field regime. The herein reported results refer to $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi)$ and $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$ for He irradiated by a dichromatic ac-field consisting of the fundamental wavelength $\lambda = 248$ nm and its 2nd, 3rd and 4th higher harmonics. The intensities are in the range 1.0×10^{12}–3.5×10^{14} W/cm^2, with the intensity of the harmonics being 1–2 orders of magnitude smaller. The calculations incorporated systematically electronic structure and electron correlation effects in the discrete and in the continuous spectrum, for ^{1}S, ^{1}P, ^{1}D, ^{1}F, ^{1}G, and ^{1}H two-electron states of even and odd parity.

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

1 Introduction

The response of atoms or molecules to dichromatic laser fields leads to the dependence of observables on the phase difference φ. This fact has been the subject of many theoretical and experimental publications. For representative analyses and results the reader is referred to [1–22]. The basic source of this dependence is the quantum mechanical interference between the various possible excitation paths. For example, the weak-field model analysis of Brumer and Shapiro [2,3] illustrated how the irradiation of a bound state with a field of commensurate frequencies, ω_1 and $\omega_2 = 3\omega_1$, may be used for the coherent phase control of the rate of production of the final products in the continuous spectrum. Additional early results based on one- electron calculations within time-dependent [6,10] and time-independent Floquet frameworks [7,8] showed the effect of φ on field-induced properties, even for strong fields. Experimental work has also revealed the effect of phase-dependent interference on atomic observables (*e.g.* [4,5,12,13,19,20]).

The situation of a polyelectronic atomic state interacting with one laser field can be treated within a time-independent or a time-dependent framework, depending on the nature of the field in connection with the atomic spectrum which is probed. In either case, one is faced, in general, with a many-electron, many-photon (MEMP) problem, whose proper solution must be achieved in the context of two regimes: one where the field is weak with respect to the state of interest, and for which lowest-order perturbation theory (LOPT) holds, and one where the field is strong and the theory and calculation must go beyond LOPT, even to infinite order.

When the polyelectronic atomic state is made to interact with a dichromatic or a polychromatic laser field, it is natural that the MEMP problem becomes very demanding, especially when one aims at the inclusion of

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all the significant electronic structure characteristics beyond the independent particle model. Apart from the ensuing many-electron problem, one has to consider the fact that there is no definition of the weak field regime, since the final rate results from the addition of amplitudes involving matrix elements with two or more fields. A time-independent, nonperturbative MEMP theory, (MEMPT), and method of computation of the energy width, $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi)$, and the energy shift, $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$, which occur when an atomic state, ground or excited, is placed in a dichromatic field, (ω_1 and ω_2), with field strengths F_1, F_2, that may be weak or strong, was developed by us recently [21,22], by extending in a practical way for commensurate frequencies, the MEMPT for the monochromatic case ([23] and references therein). The calculations in [21,22] as well as here dealt with helium for experimentally achievable situations. Specifically, we used $\omega_1 = 5$ eV ($\lambda_1 = 248$ nm) and the higher harmonics with $\omega_2 = m\omega_1$, $m = 2$, 3, 4. The intensities of the fundamental ω_1 ranged between 1.0×10^{12} and 3.5×10^{14} W/cm^2 while those of the harmonics were 1–2 orders of magnitude smaller.

The purpose of this contribution is to report results of the MEMPT for both weak and strong fields from which it follows that it is possible for the rate of multiphoton ionization as well as for the energy shift to depend in a simple way on the phase difference, φ, when the two fields with commensurate frequencies are weak. This dependence is $\cos(\varphi)$ when the harmonics are odd, and $\cos(2\varphi)$ when the harmonics are even. The nonperturbative calculations demonstrate that these rules break down when the field strengths increase. A plausible explanation of this phenomenon is given in terms of heuristic arguments.

We point out that in [21] comparison was made with the only previously available results for this case, namely those of Telnov and Chu [17b], who computed field-dressed energies only for $\varphi = 0$ and $\varphi = \pi$ by implementing a generalized Floquet formulation of the time-dependent density functional theory (TDDFT) using a Hartree-Fock functional. Our results, which include the effects of electron correlation, show the same trend as those of [17b] as a function of intensity, but differ quantitatively.

2 Heuristic argument about the form of the dependence of Γ and Δ on the phase difference

We take the dichromatic field, $F(t)$, as

$$F(t) = F_1 \cos(\omega_1 t) + F_2 \cos(\omega_2 t + \varphi), \qquad (1)$$

where $\omega_1 = \omega$, $\omega_2 = m\omega$, $m =$ even or odd integer. We ask the question whether it is possible to obtain a simple mathematical relationship between the results of the perturbation, namely Γ and Δ, and φ.

In order to facilitate the syllogism, we use as exemplar Figure 1, which shows the possibilities of multiphoton ionization for harmonic frequencies of $\omega_2 = m\omega_1$, $m = 2$, 3...7.

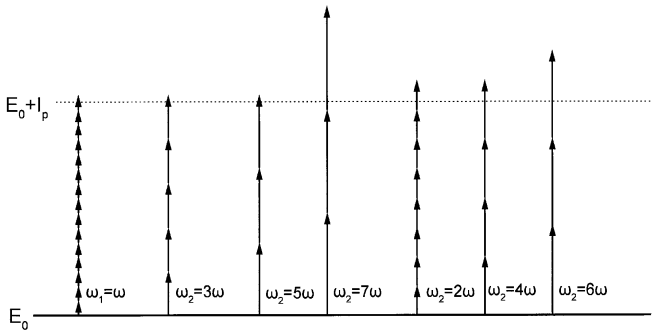

Fig. 1. Schematic diagram showing the lowest order processes for the multiphoton ionization of an atom, with ionization potential I_p, which is initially in its ground state of energy E_0. The frequencies corresponding to each process are the fundamental $\omega_1 = \omega$ and its higher harmonics $\omega_2 = m\omega$ ($m = 1, ..., 7$).

Figure 1 implies lowest order perturbative conditions, whereby both Δ and Γ are determined by the same types of matrix elements present in the lowest order perturbation theory (LOPT). The final states in the continuous spectrum have the same energy and symmetry and this leads to quantum interference of two or more paths for ω_1 and ω_2, which depends on the value of φ. We hypothesize that the intensities F_1 and F_2 of the two paths representing the maximum and the minimum number of photons necessary for ionization are such that the corresponding transition rates are about the same. This implies that $F_2 < F_1$, since the order of the process for F_1 is higher. Given this constraint, it is reasonable to expect that there must be one or more paths in between, with larger transition rates. We then assume that the dominant contribution to the partial ionization rate for the specific final state comes from intermediate paths, and that these final states are the dominant contributors to the overall interference producing the dependence of Γ and Δ on φ.

When both intensities are in the domain of LOPT, the most important final states will be those just above threshold with energies ranging from $E_0 + n_1\omega_1$ to $E_0 + n_2\omega_2$, where (n_1, n_2) are the smallest numbers of photons required for ionization by fields of frequencies ω_1 and ω_2 (Fig. 1). In order to trace the paths leading to the same final states, we can use heuristically the LOPT formulae or Figure 1, in order to substitute the path of ω_1 with the appropriate paths of $\omega_2 = m\omega$. In this way, a path consisting of steps of ω_1 with field strength F_1 (Fig. 2a) is sequentially replaced by paths of lower multiphoton order, consisting of steps of ω_1 as well as of $\omega_2 = m\omega$, with phase difference φ. We point out that, although the field Hamiltonian of the MEMPT calculations contains rotating as well as counter-rotating terms, Figure 2 does not contain all the possible ionization paths and does not indicate the possible situations whereby intermediate bound states influence the nonlinear processes. This is because we wish to simplify matters so as to be able to explore the possible effects of easily visualized paths that bring about quantum interference. So, from the properties of the

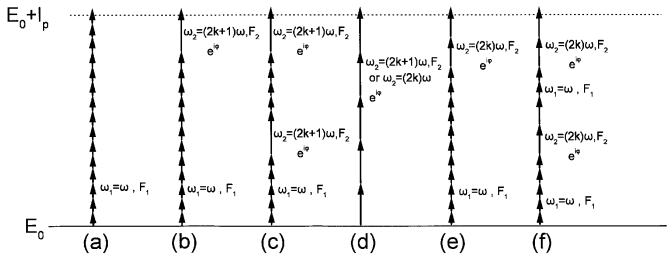

Fig. 2. Various paths for the multiphoton ionization of an atom in a dichromatic ac-field. For reasons of economy, the counter-rotating terms, which are associated with the factor $e^{-i\varphi}$, are not shown; (a) path consisting of steps of the fundamental frequency $\omega_1 = \omega$ and field strength F_1; (b) path (a) where $(2k+1)\omega$ steps are substituted with one step of F_2, $\omega_2 = (2k+1)\omega$, φ; (c) path (b) where a different set of $(2k+1)\omega$ steps is substituted with one step of F_2, $\omega_2 = (2k+1)\omega$, φ; (d) path with steps of the higher harmonic, ω_2; (e) path (a) where $(2 \times 2k)\omega$ steps are substituted with two steps of F_2, $\omega_2 = (2k)\omega$, φ; (f) path (e) where a different set of $(2 \times 2k)\omega$ steps are substituted with two steps of F_2, $\omega_2 = (2k)\omega$, φ.

generalized cross-section, we consider it plausible that the decrease of the order of the multiphoton process will result in the increase of the absolute value of the ionization probability amplitude of the corresponding path, even though F_2 is smaller than F_1. (Our numerical results confirm this trend of the ionization probability amplitudes.) At the end of this series of exchanges of paths, there must exist a path with the minimum multiphoton order, (Fig. 2d), for which the absolute value of the ionization probability amplitude is essentially equal to the one corresponding to the process of Figure 2a. This observation suggests that the paths of consecutively decreasing multiphoton order are associated with probability amplitudes of increasing absolute values, which, however, necessarily pass through a maximum and finally end near the original value. Of course, the above argument cannot be proven formally and cannot be considered as having universal validity. Nevertheless, in the present case it is in harmony with the computed final results, and it is up to future results on other systems to determine the extent of its applicability. On the other hand, we point out that, as we mention below, there are experimental results in the literature which have indeed revealed the dependence of the ionization rate on φ that results from the present arguments and computations.

We now consider two possibilities of dichromatic multiphoton ionization.

2.1 $\omega_2 = m\omega = (2k+1)\omega$

The replacement of $2k+1$ steps of $\omega_1 = \omega$, with *one* step of $\omega_2 = (2k+1)\omega$, (see the path of Fig. 2b), results in a subspace of final states of the same energy and symmetry (even or odd parity). Actually, the lower the order of the multiphoton ionization is, the smaller is the subspace of the final states as regards their total angular momentum eigenstates. For example, in He, for the process of Figure 2a the final state subspace consists of continuum states of symmetry ^1L with $L = n_1, n_1-2, n_1-4, ...$ and for the process of Figure 2b the corresponding symmetry is ^1L with $L = n_1-2k, n_1-2k-2, n_1-2k-4, ...$ The next path (Fig. 2c) is constructed by replacing another group of $2k+1$ steps of ω_1 with *one* of (ω_2, F_2, φ), and so on.

The total ionization rate $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi)$ is obtained from the square of the absolute value of the sum of the probability amplitudes of the various paths to the same final states whose energies range from $E_0 + n_1\omega_1$ to $E_0 + n_2\omega_2$ and whose parities may be even or odd. We write:

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k+1)\omega, F_2, \varphi) \sim \left| \sum_p Q_p \, e^{ip\varphi} \right|^2 + ... \quad (2)$$

where p changes in steps of 1 and Q_p are the ionization probability amplitudes of each path leading to the same final state. The remaining terms in (2) represent summations of other amplitudes leading to other final states. Each state is characterized by its energy and its symmetry. The final states that contribute significantly to Γ and give rise to quantum interference could be more than one. For each such final state, one of the Q_p's represents the maximum value (Q_M). With the heuristic assumption that, for not too low frequencies, Q_M and $Q_{M\pm1}$ are the leading terms in each factor of equation (2), (with phase difference φ), the quantum interference is driven by two terms:

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k+1)\omega, F_2, \varphi) \sim A + B\cos(\varphi) + ... \quad (3)$$

With increasing field strengths, the higher terms of equation (3) should come into play.

The form (3) and the dependence of $\Gamma(\Delta)$ on $\cos(\varphi)$ for weak fields, and the breakdown when the field strengths increase, is verified by the nonperturbative MEMPT calculations reported in [21,22] and here. Furthermore, confirmation of this form can be found in already published observations of a rather different system. Specifically, Chen and Elliot [5] deduced such a cosine dependence for the particular case of their pioneering $(\omega, 3\omega)$ experiments on the $6s\ ^1\mathrm{S} \rightarrow 6p\ ^1\mathrm{P}^0$ transition in atomic mercury with laser pulses consisting of an intense component of $\lambda_1 = 554$ nm and a weak component of $\lambda_2 = 185$ nm.

2.2 $\omega_2 = m\omega = (2k)\omega$

Unlike Section 2.1, the replacement of $2k$ steps of (ω_1, F_1) with one step of (ω_2, F_2, φ) does not result in a subspace of final states of the same parity. Instead, it is the replacement of $2(2k)$ steps (of ω_1, F_1) by *two* steps of (ω_2, F_2, φ) that gives a path which ends in a subspace of final states of the same energy and parity (Fig. 2e). The next path, depicted in Figure 2f, is obtained by replacing another group of $2(2k)$ steps of (ω_1, F_1) with *two* steps of (ω_2, F_2, φ), and

so on. Similarly to the Section 2.1, the total ionization rate is given by

$$\Gamma(\omega_1=\omega,F_1;\omega_2=(2k)\omega,F_2,\varphi) \sim \left|\sum_p Q_{2p}\,\mathrm{e}^{\mathrm{i}2p\varphi}\right|^2+... \quad (4)$$

Following the same arguments as before, (Sect. 2.1), we conclude that Γ is driven by two terms:

$$\Gamma(\omega_1=\omega,F_1;\omega_2=(2k)\omega,F_2,\varphi) \sim A+B\cos(2\varphi)+... \quad (5a)$$
$$= A'+B'\cos^2(\varphi)+... \quad (5b)$$

Again, the higher order terms of equation (5) should acquire significant values with increasing field strengths.

The form (5) and its breakdown as the field strength increases is verified by our MEMPT computations. Furthermore, it has been observed in the past, without however having been subjected to analysis, in larger atoms as well. Specifically, sometime ago, Szöke, Kulander and Bardsley [6] obtained it by fitting the measurements of Muller *et al.* [4] on Kr, where Kr was irradiated by a dichromatic field with $\lambda_1 = 1064$ nm and its 2nd harmonic $\lambda_2 = 532$ nm.

3 Extended MEMPT and application to He for weak and strong dichromatic ac-fields of $\lambda = 248$ nm ($\omega_1 = \omega = 5$ eV) and its higher harmonics ($\omega_2 = m\omega, m = 2, 3, 4$)

The theory of field induced interference in multiphoton ionization presented here, has been supported by quantitative results on He, for experimentally verifiable conditions. It is noteworthy that the present work has dealt not only with the behavior of the transition rates, a subject which is at the focus of research on coherent control, but also with that of the energy shift, Δ, and its dependence on the phase difference. Since Δ can, under certain conditions, be related to measurable nonlinear dynamic polarizabilities, our results provide an additional opportunity for experimental exploration.

Since the formalism and the methods of the MEMPT for atoms in polychromatic fields with commensurate frequencies was given recently elsewhere, together with the description of the application to He [21,22], we do not repeat them here. However, we should say a few words about essential points of the MEMPT, whose initial presentation was made in the late 1980s, with applications to the negative ions of H and Li, treated as polyelectronic systems and not as one electron models [23–25]. (See also the recent work on multiphoton detachment of H^- [26].) The response of He to a monochromatic ac-field was treated recently, within the MEMPT, in [27].

As discussed in [21–27], the physics of multiphoton ionization induced by a cycle-averaged interaction of an ac-field with an atom, can be formulated in terms of a state-specific complex eigenvalue Schrödinger equation, whose eigenfunction consists of two parts that are represented by different types of function spaces. Their distinct physical content and the complex eigenvalue emerge naturally from an argument based on configuration-interaction between the field-dressed discrete state and the ionized continuum and consideration of the asymptotic form under resonance boundary conditions [28]. This asymptotic form expresses an outgoing wave with complex energy, (Gamow orbital), which is not square-integrable. As a result, the corresponding matrix elements are divergent. However, these can be regularized even for the electric field linear potential with the help of the Dykhne-Chaplik transformation $\rho = \mathrm{e}^{\mathrm{i}\theta}$ on the coordinate of the Gamow orbital [28]. In this way, the sought-after solution becomes square-integrable ($\mathcal{L}^2$) but the problem is now non-Hermitian.

The $\mathcal{L}^2$ solution, $\Psi_0(\rho)$, which is connected adiabatically to the initial unperturbed atomic state, Ψ_0, is expressed formally as

$$|\Psi_0(\rho)\rangle = \sum_{i,n}\alpha_{i,n}(\theta)|\psi_i(\rho);n\rangle + \sum_{j,n} b_{j,n}(\theta)|X_j(\rho);n\rangle \quad (6)$$

where ψ_i denotes bound states and the localized parts of the autoionizing states, X_j denotes the $\mathcal{L}^2$ "scattering" states and n is the index for the photon states. In the semiclassical approximation, when the external field is a periodic function of time (ac-field), the Fourier analysis of the time-dependent wave-functions leads to terms with index n, to which we attach the name "photon states". Expansion (6) is the same as the one used in the monochromatic case since the frequencies of the dichromatic (or polychromatic in the general case) field are commensurate. The only photon states used, which are necessary for the accurate description of $\Psi_0(\rho)$, are those of the fundamental frequency. In the general polychromatic case with incommensurable frequencies, it would be necessary to use for each frequency separate photon states, a fact which increases considerably the numerical load toward the solution of the MEMP problem.

According to the MEMPT, the dressed Hamiltonian coordinates are real and not complex, as they are in the Floquet, complex scaling methods [17]. The emphasis is on representing each significant state of the spectrum by a state-specific wavefunction in expansion (6), together with other terms representing virtual states and the continuous spectrum. The correlated function space used in this work was the one that was presented in [21,27]. Therefore, we do not repeat it here. We simply note that the space of X_j contains $\mathcal{L}^2$ two-electron functions of the form $1s\otimes\varepsilon\ell$, where the orbital $\varepsilon\ell$ is expanded in terms of Slater type orbitals (STO) with a complex coordinate:

$$\xi_k(\rho^*) = \xi_k(r\mathrm{e}^{-\mathrm{i}\theta}) = C_k(\theta)r^k\,\mathrm{e}^{-\alpha\rho^*}. \quad (7)$$

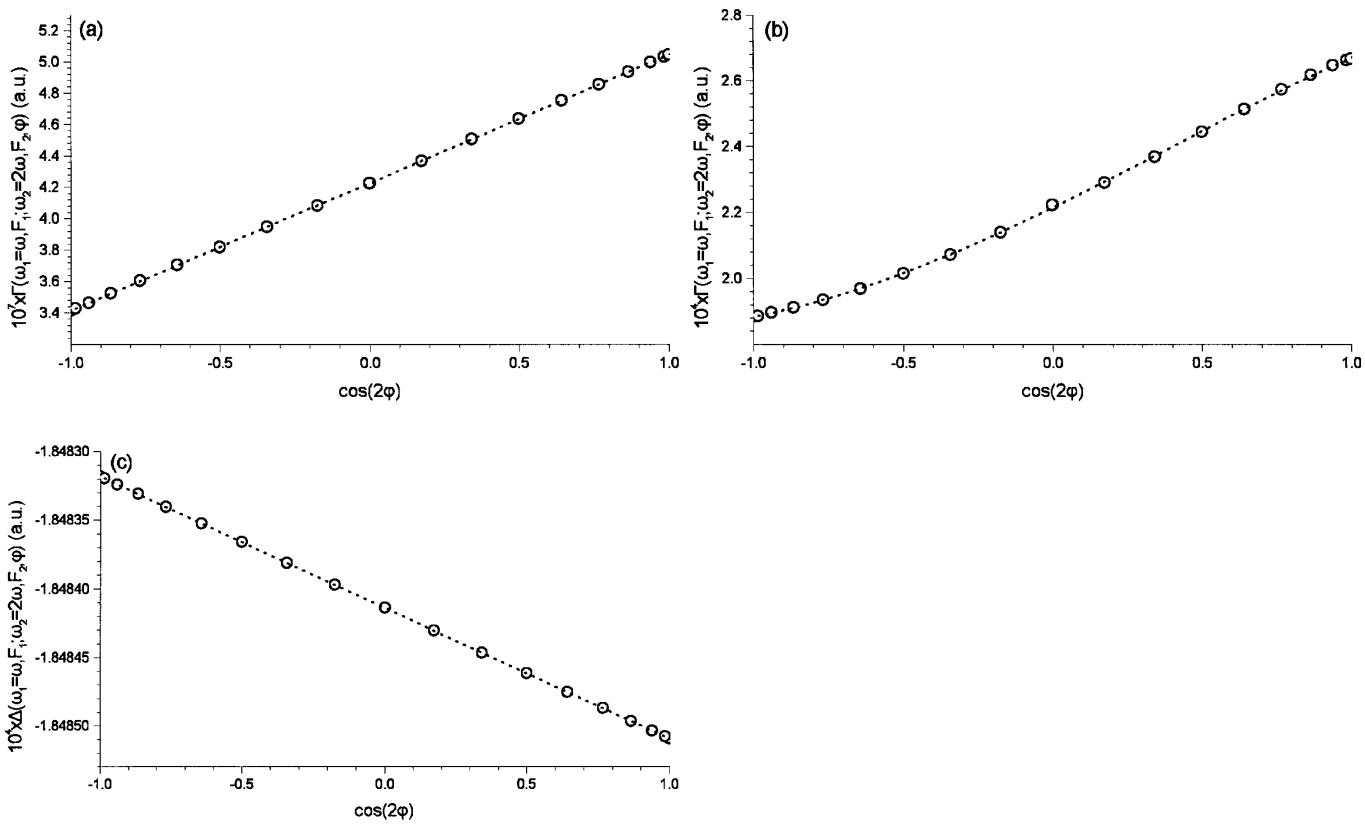

Fig. 3. (a) Multiphoton ionization rates (Γ in a.u. – open circles) of the He ground state irradiated by linearly polarized field of frequency $\omega = 0.18373$ a.u. ($\lambda = 248$ nm) with intensity $I_1 = 4.5 \times 10^{13}$ W/cm^2 and by its second harmonic ($\omega_2 = 2\omega$) with $I_2 = 6.1 \times 10^{12}$ W/cm^2, as a function of cos(2φ). The dotted line represents the fitted polynomial of equation (8). (b) As in (a), but for $I_1 = 3.5 \times 10^{14}$ W/cm^2 and $I_2 = 3.2 \times 10^{13}$ W/cm^2. (c) Energy shift (Δ in a.u. – open circles) of the He ground state irradiated by linearly polarized field of frequency $\omega = 0.18373$ a.u. ($\lambda = 248$ nm) with intensity $I_1 = 1.4 \times 10^{13}$ W/cm^2 and by its second harmonic ($\omega_2 = 2\omega$) with $I_2 = 3.5 \times 10^{12}$ W/cm^2, as a function of cos(2φ). The dotted line represents the fitted polynomial of equation (8).

For each ℓ ($\ell = 0, ..., 5$) the continuum orbitals were expanded in $10\,\xi_k(\rho^*)$, except for $\ell = 5$, for which $8\,\xi_k(\rho^*)$ were used.

The convergence of the calculation is achieved when, for a reasonable range of the values of the parameters present in the basis set and of the number of the photon states, the complex eigenvalue remains essentially unchanged. Specifically, it was found that convergence was established when the number of the photon states reached ten or eleven, and when the parameters in (7) are: $\alpha = 1.5$ and $\theta = 0.3$ rad.

Following the arguments of Section 2, we classify the results with respect to the order of the harmonics.

3.1 Even harmonics ($\omega_2 = (2k)\omega$)

3.1.1 $\omega_2 = 2\omega$

Figure 3a shows the ionization rates of the He ground state irradiated by linearly polarized field of frequency $\omega = 0.18373$ a.u. ($\lambda = 248$ nm) with intensity $I_1 = 4.5 \times 10^{13}$ W/cm^2 and by its second harmonic with $I_2 = 6.1 \times 10^{12}$ W/cm^2, as a function of cos(2φ). Even though the MEMPT calculation is nonperturbative, what has emerged is a very good linear dependence of the rate on cos(2φ), in accordance with the prediction given in Section 2. The dotted line in Figure 3a represents the fitted polynomial in powers of cos(2φ), to the *ab initio* rates:

$$\Gamma = A + B_1 \cos(2\varphi) + B_2 \cos^2(2\varphi) + ..., \qquad (8)$$

with $A = 4.23 \times 10^{-7}$ a.u., $B_1 = 8.16 \times 10^{-8}$ a.u., $B_2 = 4.2 \times 10^{-10}$ a.u. The coefficient of the quadratic term is more than two orders of magnitude smaller than B_1. On the other hand, for $I_1 = 3.5 \times 10^{14}$ W/cm^2 and $I_2 = 3.2 \times 10^{13}$ W/cm^2, the rates deviate from the linear dependence on cos(2φ) (Fig. 3b). The fitted polynomial of equation (8) (dotted line in Fig. 3b) yields the coefficients: $A = 2.2 \times 10^{-4}$ a.u., $B_1 = 4.3 \times 10^{-5}$ a.u., $B_2 = 7.0 \times 10^{-6}$ a.u. In this case, the coefficient B_2 is less than an order of magnitude smaller than B_1.

In addition, for intensities $I_1 = 1.4 \times 10^{13}$ W/cm^2 and $I_2 = 3.5 \times 10^{12}$ W/cm^2, the energy shift $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$ varies linearly with cos(2φ), as shown in Figure 3c, with the dotted line representing a fitted polynomial similar to the one of the right hand side of equation (8). For higher intensities the energy shift

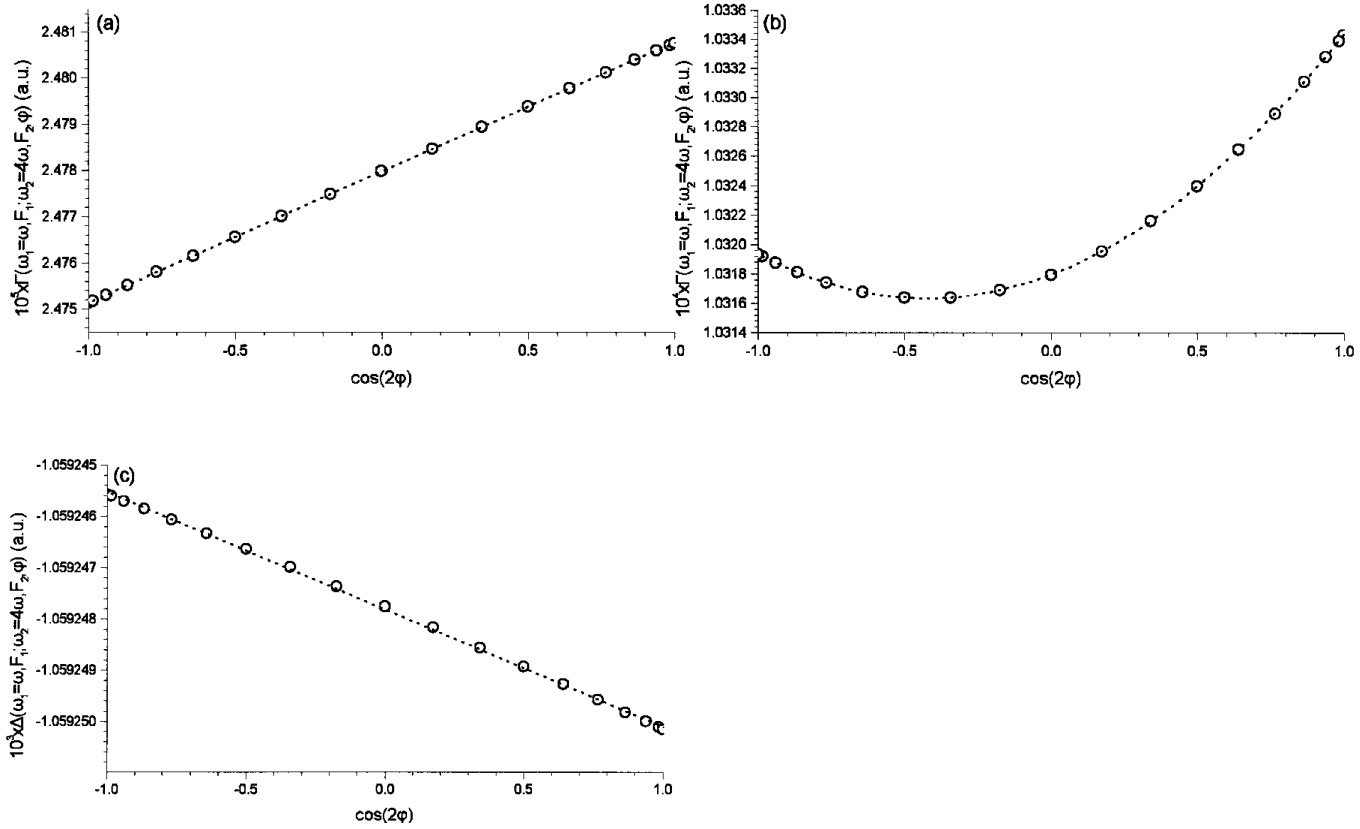

Fig. 4. (a) As in Figure 3a, but for $\omega_2 = 4\omega$, $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2. (b) As in (a), but for $I_1 = 2.24 \times 10^{14}$ W/cm^2 and $I_2 = 3.5 \times 10^{12}$ W/cm^2. (c) As in Figure 3c, but for $\omega_2 = 4\omega$, $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2.

deviates from the linear dependence on $\cos(2\varphi)$, as in the case of the rates. The slope of the straight line (coefficient B_1) should be connected to a certain component of a high order dynamic hyperpolarizability tensor. A more elaborate treatment of these quantities will be given, hopefully, in a future publication.

3.1.2 $\omega_2 = 4\omega$

Figure 4a shows the ionization rate of He for $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2, as a function of $\cos(2\varphi)$. Again, a linear dependence is obtained. What is interesting is that this linear dependence persists even when the intensity I_1 for the fundamental frequency is beyond the domain of the validity of the LOPT. (For a monochromatic Γ with $\omega = 5$ eV, LOPT breaks down around $I = 7 \times 10^{13}$ W/cm^2. For weak intensities, five 5 eV photons are needed for ionization of He. However, for $I_1 = 1 \times 10^{14}$ W/cm^2, six photons are required, due to the ponderomotive shift.) The coefficients of the fitted polynomial (dotted line in Fig. 4a) are now $A = 2.48 \times 10^{-5}$ a.u., $B_1 = 2.83 \times 10^{-8}$ a.u., $B_2 = -2.63 \times 10^{-10}$ a.u. By increasing the intensities to $I_1 = 2.24 \times 10^{14}$ W/cm^2 and $I_2 = 3.5 \times 10^{12}$ W/cm^2, a deviation from the linear dependence is obtained, as it is shown in Figure 4b. The values of the coefficients of the fitted polynomial (dotted line) are: $A = 1.03 \times 10^{-4}$ a.u., $B_1 = 7.62 \times 10^{-8}$ a.u., $B_2 = 8.8 \times 10^{-8}$ a.u.

As in the previous case and for intensities $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2, the energy shift $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$ varies linearly with $\cos(2\varphi)$, as shown in Figure 4c. Similarly, the dotted line represents the fitted polynomial of equation (8) and for higher intensities the energy shift deviates from the linear dependence on $\cos(2\varphi)$.

3.2 Odd harmonics ($\omega_2 = (2k+1)\omega$)

3.2.1 $\omega_2 = 3\omega$

Figure 5a shows the rate of He for $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2, as a function of $\cos(\varphi)$. As in Figure 4a, the rate is governed by a linear dependence on $\cos(\varphi)$, even though the intensity I_1 is beyond the domain of the validity of the LOPT for the monochromatic case. The coefficients of the fitted polynomial (dotted line)

$$\Gamma = A + B_1 \cos(\varphi) + B_2 \cos^2(\varphi) + ... \qquad (9)$$

are, $A = 2.75 \times 10^{-6}$ a.u., $B_1 = 1.56 \times 10^{-6}$ a.u., $B_2 = 5.82 \times 10^{-8}$ a.u.

An increase of the intensities to $I_1 = 1.7 \times 10^{14}$ W/cm^2 and $I_2 = 1.4 \times 10^{13}$ W/cm^2 results in a dependence of the

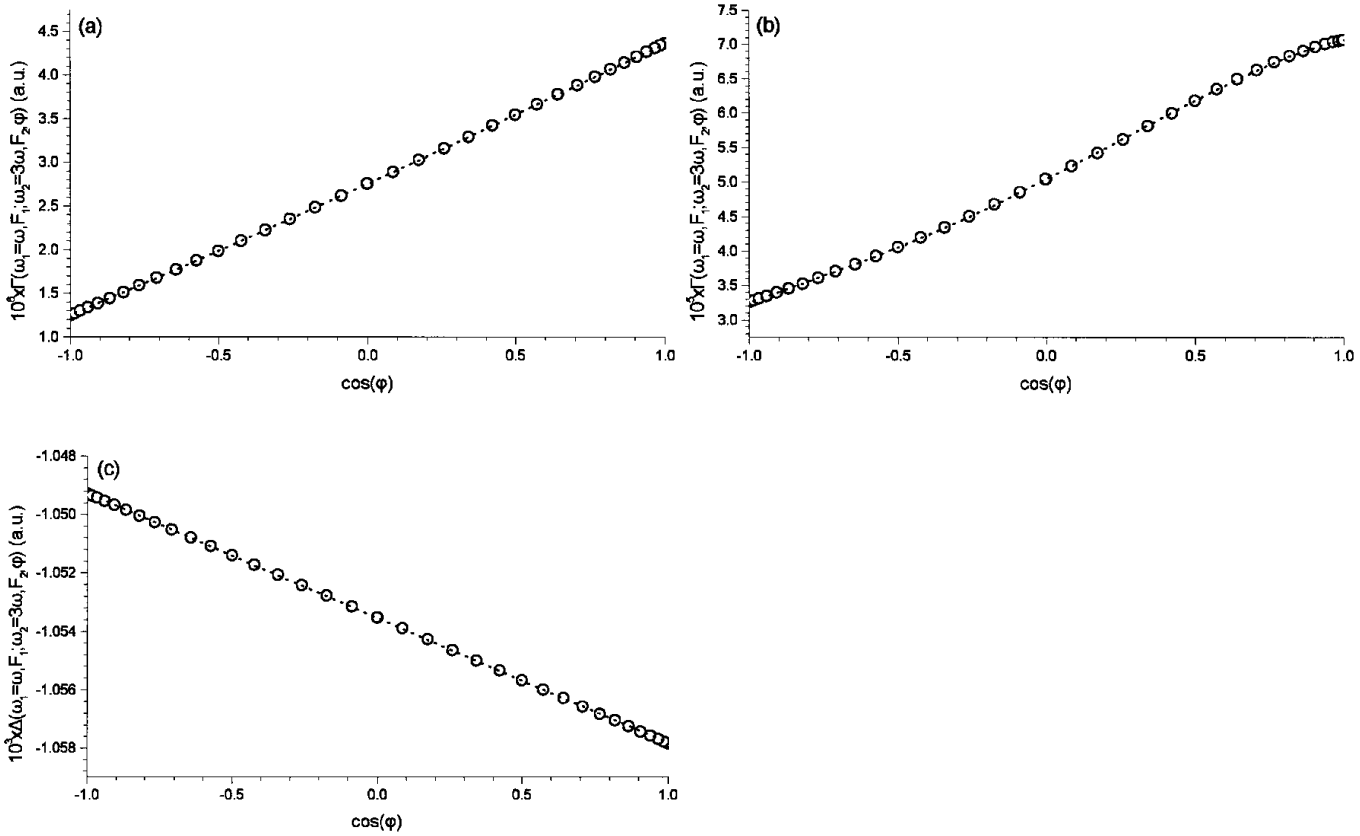

Fig. 5. (a) Multiphoton ionization rates (Γ in a.u. – open circles) of the He ground state irradiated by linearly polarized field of frequency $\omega = 0.18373$ a.u. ($\lambda = 248$ nm) with intensity $I_1 = 1 \times 10^{14}$ W/cm^2 and by its third harmonic ($\omega_2 = 3\omega$) with $I_2 = 1 \times 10^{12}$ W/cm^2, as a function of cos(φ). The dotted line represents the fitted polynomial of equation (9). (b) As in (a), but for $I_1 = 1.7 \times 10^{14}$ W/cm^2 and $I_2 = 1.4 \times 10^{13}$ W/cm^2. (c) As in Figure 4c, but for $\omega_2 = 3\omega$, $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2.

rate which is not linear with respect to cos(φ). This is shown in Figure 5b. When we fit the polynomial of equation (9) to the MEMPT values, the values of the coefficients are $A = 5.04 \times 10^{-5}$ a.u., $B_1 = 2.2 \times 10^{-5}$ a.u., $B_2 = 3.8 \times 10^{-6}$ a.u.

As in the previous cases and for intensities $I_1 = 1 \times 10^{14}$ W/cm^2 and $I_2 = 1 \times 10^{12}$ W/cm^2, the energy shift $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi)$ varies linearly with cos(φ), as shown in Figure 5c, with the dotted line representing the fitted polynomial of equation (9). Again, the higher the intensities are, the stronger the deviation from the linear dependence of the energy shift is.

4 Conclusion

The analysis and the quantitative MEMPT results for He presented in this paper indicate that when atoms are irradiated by weak dichromatic ac-fields ($F_1, \omega_1 = \omega; F_2, \omega_2 = m\omega, \varphi$), path interference is such that the multiphoton ionization rate and the energy shift are governed by a simple rule as regards their relation to the phase difference, φ. Indeed, for odd ($m = 2k+1$) higher harmonics, both the field induced energy width and shift vary linearly with cos(φ), whilst for even higher harmonics they vary linearly with cos(2φ). This behavior provides an index for defining the domain of weak fields in the dichromatic case, just like the dependence of the rate on I^n, as it is clearly derived from the LOPT, provides the index for weak fields in the monochromatic case. Given this distinction, one may define the strong field regime for the dichromatic case for spectra such as those of He and other noble gases, as that combination of the two intensities whereby the dependence of the multiphoton ionization rate on φ is no longer simple, since terms with higher powers of cos(φ) and cos(2φ) contribute significantly.

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