

He in dichromatic weak or strong ac fields of $\lambda_1=248$ nm and $\lambda_2=(1/m) 248$ nm ($m=2,3,4$)

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We have computed multiphoton ionization rates for He irradiated by a dichromatic ac field consisting of the fundamental wavelength $\lambda = 248$ nm and its second-, third-, and fourth-higher harmonics. The intensities are in the range $1.0 \times 10^{12} - 3.5 \times 10^{14}$ W/cm², 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 ¹S, ¹P, ¹D, ¹F, ¹G, and ¹H two-electron states of even and odd parity. They were done by implementing a time-independent, nonperturbative many-electron, many-photon theory which obtains cycle-averaged complex eigenvalues, whose real part gives the field-induced energy shift, $\Delta(\omega_1, F_1; \omega_2, F_2, \varphi_2)$, and the imaginary part is the multiphoton ionization rate, $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi_2)$, where ω is the frequency, F is the field strength, and φ_2 is the phase difference. Through analysis and computation we show that, provided the intensities are weak, the dependence of $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi_2)$ on φ_2 is simple. Specifically, for odd higher harmonics, Γ varies linearly with $\cos(\varphi_2)$ whilst for even higher harmonics it varies linearly with $\cos(2\varphi_2)$. 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 $\cos(\varphi_2)$ and $\cos(2\varphi_2)$ become important, these rules break down and we reach the strong-field regime.

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I. INTRODUCTION

In a recent publication [1], we reported the results of many-electron, many-photon theory (MEMPT) computations of the cycle-averaged multiphoton ionization rates (MPIR) and linear and nonlinear polarizabilities of He interacting with a linearly polarized ac field for frequencies in the range $\omega = 4.9 - 26.4$ eV. In this paper we extend the implementation of the MEMPT to the dichromatic case ($\omega_1 = \omega = 5$ eV or $\lambda_1 = 248$ nm) with commensurate frequencies of higher harmonics, $\lambda_2 = (1/m)248$ nm, $m = 2, 3, 4$, and provide reliable MPIRs which can be tested experimentally. The intensities are in the range $1.0 \times 10^{12} - 3.5 \times 10^{14}$ W/cm², with the intensity of the harmonics being 1–2 orders of magnitude smaller. Obviously, the MPIRs depend on intensity. Furthermore, they depend on the phase difference φ_2 between the two fields, and the present study has produced unique results not only for the particular system but also for the phenomenology of the problem, which should be applicable to multiphoton processes in other systems as well.

The fact that the response of atoms or molecules to dichromatic laser fields leads to the dependence of observables on φ_2 has been the subject of many theoretical and experimental publications. For representative analyses and results the reader is referred to Refs. [2–21]. 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 [3,4] 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 [7,11] and time-independent Floquet frameworks [8,9] showed the effect of φ_2 on field-induced properties, even for strong fields. Experimental work has also revealed the effect of phase-dependent interference on atomic observables [e.g., 5, 6, 13, 14, 20, 21].

The work described below was carried out by combining heuristic arguments with MEMPT all-order calculations. The former was applied in order to explore whether it is possible for the MPIR to be connected in a simple way to the phase difference when the intensities of the two interfering waves are weak. The latter (MEMPT) was applied in order to produce accurate results for weak as well as for strong fields, which, not only should be useful as such, but they could provide quantitative information about the degree of validity of the heuristic arguments. Indeed, the results show that, at least for He, and probably for other systems where similar conditions are met, there is an underlying simple “law” connecting the MPIRs with the phase difference φ_2 .

II. HEURISTIC ARGUMENTS FOR THE FORM OF THE DEPENDENCE OF THE MPIR ON THE PHASE DIFFERENCE

Let us consider a dichromatic field $F(t)$ in the form

$$F(t) = F_1 \cos(\omega_1 t + \varphi_1) + F_2 \cos(\omega_2 t + \varphi_2), \quad (1)$$

where $\varphi_1 = 0$ (without loss of generality for the present argument), and $\omega_1 = \omega$, $\omega_2 = m\omega$, and m is an even or odd

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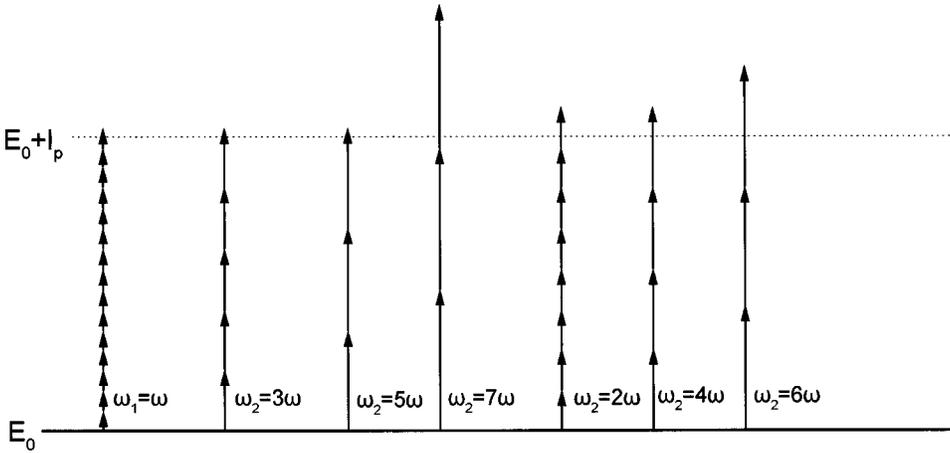


FIG. 1. Schematic diagram showing the lowest-order processes for the multiphoton ionization of an atom with ionization potential I_p , which is initially in its ground state of energy E_0 . The frequencies corresponding to each process are the fundamental $\omega_1 = \omega$ and its higher harmonics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

integer. We ask the question whether it is possible to obtain the relationship between the rate and φ_2 in terms of a simple function. In order to facilitate the syllogism, we take as an example Fig. 1, which shows the possibilities of multiphoton ionization for harmonic frequencies of $\omega_2 = n\omega$, $n = 2, 3, \dots, 7$.

The quantum interference of two or more paths for ω_1 and ω_2 to final states with the same energy and symmetry brings about a dependence of the total rate on φ_2 . We assume that for such final states, the individual intensity for each path is weak, [the lowest-order perturbation theory (LOPT) expression is applicable], and that for each final ionized state for which there is one path with the maximum number of required photons and one path with the minimum such number, the transition rates via these two paths are about the same. The second assumption leads to the conclusion that the dominant contribution to the partial rate for ionization to the specific final state comes from paths of order between those corresponding to the minimum and the maximum number of required photons. These specific final states are also assumed to be the dominant contributors to the overall interference which gives the dependence of the total rate on φ_2 . When these assumptions are met, it means that $F_2 \ll F_1$, since the multiphoton process for the fundamental frequency is of higher order.

When both intensities are in the domain of LOPT, we expect that 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 formulas or figures such as Fig. 1 to substitute the path of ω_1 with the appropriate paths of $\omega_2 = n\omega$. In this way, a path consisting of steps of ω_1 with field strength F_1 [Fig. 2(a)] is sequentially replaced by paths of lower multiphoton order, consisting of steps of ω_1 as well as of $\omega_2 = n\omega$, with field strengths F_1 and F_2 and of phase difference φ_2 . From the properties of the generalized cross section, we consider it plausible that the decrease of the order of the multiphoton process for independent paths will result in the increase of the absolute value of the ionization probability amplitude of the corresponding path, provided a suitable combination of (F_1, F_2) exists, even for cases where F_2 is smaller than F_1 . (Our numerical results confirm this trend of the ionization probability amplitudes.) Furthermore, at the end of this series of exchanges of paths, we will find the path with the minimum multiphoton order [i.e., Fig. 2(d)], whose absolute value of

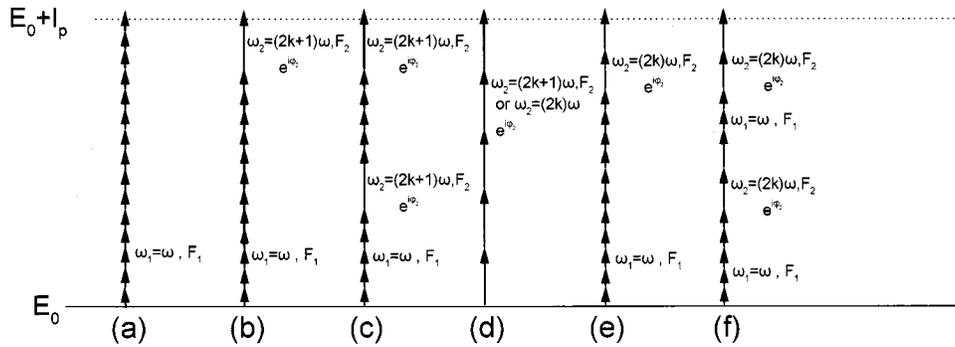


FIG. 2. Various paths for the multiphoton ionization of an atom in a dichromatic ac-field. (a) Path consisting of steps of the fundamental frequency $\omega_1 = \omega$ and field strength F_1 . (b) Path (a) where $2k + 1 \omega$ steps are substituted with one step of F_2 , $\omega_2 = (2k + 1)\omega$, φ_2 . (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_2 . (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_2 . (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_2 .

the ionization probability amplitude is essentially equal to the one corresponding to the process of Fig. 2(a). 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. On the other hand, we are aware of the fact that the argument about the increase of the ionization probability with decreasing order of the multiphoton process, cannot be considered general. In other words, there may be cases where the absolute values of the probability amplitudes associated with paths of decreasing multiphoton order would pass through a broad maximum or would be of similar order of magnitude

Keeping the above arguments in mind, we proceed by considering two cases of dichromatic multiphoton ionization.

(i) $\omega_2 = m\omega = (2k+1)\omega$. The replacement of $2k+1$ steps of $\omega_1 = \omega$, with *one* step of $\omega_2 = (2k+1)\omega$ [see the path of Fig. 2(b)], 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 Fig. 2(a), the final-state subspace consists of continuum states of symmetry 1L with $L = n_1, n_1 - 2, n_1 - 4, \dots$ and for the process of Fig. 2(b) the corresponding symmetry is 1L with $L = n_1 - 2k, n_1 - 2k - 2, n_1 - 2k - 4, \dots$. The next path [see Fig. 2(c)] is constructed by replacing another group of $2k+1$ steps of ω_1 with *one* of $(\omega_2, F_2, \varphi_2)$, and so on.

The total ionization rate $\Gamma(\omega_1, F_1; \omega_2, F_2, \varphi_2)$ 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

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k+1)\omega, F_2, \varphi_2) \sim \left| \sum_p Q_p e^{ip\varphi_2} \right|^2 + \dots, \quad (2)$$

where p changes in steps of 1 and Q_p are the ionization probability amplitudes of each path. Each additive factor of the right-hand side of Eq. (2) consists of a summation of terms of different orders of magnitude. Indeed, as we mentioned before, one of the Q_p 's represents the maximum value (Q_M). With the heuristic assumption that Q_M and $Q_{M\pm 1}$ are the leading terms in each factor of Eq. (2), the quantum interference is driven by two terms

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k+1)\omega, F_2, \varphi_2) \sim A + B \cos(\varphi_2) + \dots \quad (3)$$

With increasing field strengths, the higher terms of Eq. (3) should come into play. In the cases where the Q_p 's pass through a broad maximum or are of similar order of magnitude, the higher terms of Eq. (3) acquire significant values even for weak-field strengths. We point out that Chen and Elliot [6] deduced a cosine dependence of the type (3) for the particular case of their pioneering $(\omega, 3\omega)$ experiments on the $6s \ ^1S \rightarrow 6p \ ^1P^o$ 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.

(ii) $\omega_2 = m\omega = (2k)\omega$. Unlike case (i), the replacement of $2k$ steps of (ω_1, F_1) with one step of $(\omega_2, F_2, \varphi_2)$ 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 $(\omega_2, F_2, \varphi_2)$ that gives a path which ends in a subspace of final states of the same energy and parity [see Fig. 2(e)]. The next path, depicted in Fig. 2(f), is obtained by replacing another group of $2(2k)$ steps of (ω_1, F_1) with two steps of $(\omega_2, F_2, \varphi_2)$, and so on. Similarly to the case (i), the total ionization rate is given by

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k)\omega, F_2, \varphi_2) \sim \left| \sum_p Q_{2p} e^{i2p\varphi_2} \right|^2 + \dots \quad (4)$$

Following the same arguments as before [case (i)], we conclude that the MPIR is driven by two terms:

$$\Gamma(\omega_1 = \omega, F_1; \omega_2 = (2k)\omega, F_2, \varphi_2) \sim A + B \cos(2\varphi_2) + \dots \quad (5a)$$

$$= A' + B' \cos^2(\varphi_2) + \dots \quad (5b)$$

Again, the higher-order terms of Eq. (5) should acquire significant values with increasing field strengths. This is indeed found from our computations, whose results are presented in the following section. In the cases where the Q_p 's pass through a broad maximum or are of similar order of magnitude, the higher terms of Eq. (5a) acquire significant values even for weak-field strengths. We point out that the dependence of the type (5) was reported by Szöke, Kulander, and Bardsley [7], who obtained it by simply fitting the measurements of Muller *et al.* [5], where Kr was irradiated by a dichromatic field with $\lambda_1 = 1064$ nm and its second harmonic $\lambda_2 = 532$ nm.

III. POLYCHROMATIC MEMPT AND APPLICATION TO HE FOR WEAK AND STRONG FIELDS

We now turn to the calculation from first principles of the MPIRs of He interacting with polychromatic fields. The calculations are quantitative since they incorporate the effects of electronic structure and of correlations. In order to do this we adapt the MEMPT to the case where the external ac field is a superposition of a linearly polarized field and its harmonics:

$$F(t) = \sum_{m=1}^{N_c} F_m \cos(\omega_m t + \varphi_m), \quad (6)$$

where ω_m is the integer multiple of the fundamental frequency ω .

As discussed in [1,22,23], the physics of multiphoton ionization induced by a constant 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. 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 $\rho = e^{i\theta}$ on the coordinate of the Gamow orbital [24]. In this way, the sought-after solution becomes square integrable (\mathcal{L}^2) but the problem is now non-Hermitian.

By generalizing the MEMPT to the case of the field of Eq. (6), the atom plus field Hamiltonian in the linearly polarized electric-dipole approximation has the form

$$H = H_{\text{atom}} + \sum_{m=1}^{N_c} \hbar \omega_m \alpha_{\omega_m}^\dagger \alpha_{\omega_m} - \frac{1}{2} \sum_{m=1}^{N_c} F_m z (e^{i\varphi_m} \alpha_{\omega_m}^\dagger + e^{-i\varphi_m} \alpha_{\omega_m}), \quad (7)$$

where $\alpha_{\omega_m}^\dagger$ and α_{ω_m} are the photon creation and annihilation operators.

The complex eigenvalue can be written as

$$z_0 = E_0 + \Delta(\omega_1, F_1; \omega_2, F_2, \varphi_2) - \frac{1}{2} i \Gamma(\omega_1, F_1; \omega_2, F_2, \varphi_2), \quad (8)$$

where E_0 is the free atomic state energy, Δ is the energy shift, and Γ is the MPIR.

Following earlier work on the complex eigenvalue Schrödinger equation for field-free resonance states, we have advocated that the many-electron, many-photon problem is simplified in a practical way by aiming at the separation of the function space representing the electronic structure and correlation effects contributing to the initial bound states, from the contribution of the scattering functions, which interact with the bound states via the external field of Eq. (6) and cause the energy shift Δ and width Γ . This separation is crucial for the solution to all orders of the problem of computing properties of many-electron atoms in strong ac fields. Formally, the \mathcal{L}^2 solution $\Psi_0(\rho)$, which is connected adiabatically to the initial unperturbed atomic state ψ_0 , is expressed as

$$|\Psi_0(\rho)\rangle = \sum_{i,n} \alpha_{i,n}(\theta) |\psi_i(\rho); n\rangle + \sum_{j,n} b_{j,n}(\theta) |X_j(\rho); n\rangle, \quad (9)$$

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. Expansion (9) is the same as the one used in the monochromatic case [$N_c = 1$ in Eq. (6)], since the frequencies of the dichromatic (or polychromatic in the general case) field are commensurate. The only photon states used are those of the fundamental frequency. Otherwise, it would be necessary to use for each

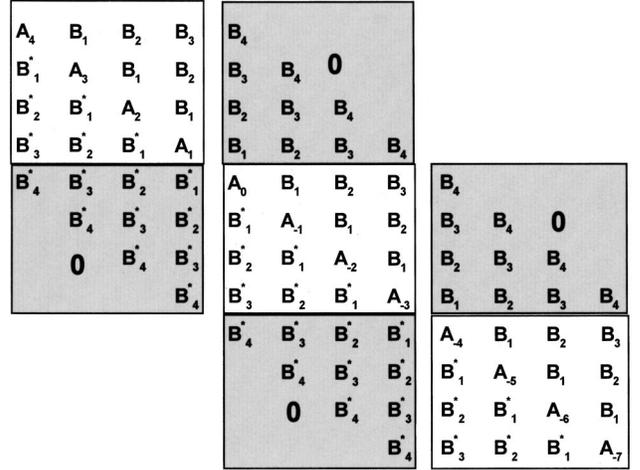


FIG. 3. The atom-field polychromatic Hamiltonian matrix \mathbf{H}_F (see text) is separated appropriately (white and gray areas), so as to permit the formulation of the corresponding eigenvalue problem in a manner similar to that of the monochromatic case previously solved.

frequency separate photon states, a fact which increases considerably the numerical load toward the solution of the MEMP problem.

Using the many-electron, many-photon basis set of Eq. (9), we construct the state-specific matrix equation

$$\mathbf{H}_F \mathbf{x} = z_0 \mathbf{x} \quad (10)$$

where the structure of \mathbf{H}_F is shown in Fig. 3. The eigenvector \mathbf{x} consists of the coefficients $\alpha_{i,n}(\theta)$ and $b_{j,n}(\theta)$ of Eq. (9). The building blocks of \mathbf{H}_F shown in Fig. 3 are

$$\mathbf{A}_n = \mathbf{A} + n \omega \mathbf{I}, \quad (11a)$$

$$\mathbf{B}_m = F_m \mathbf{V} e^{i\varphi_m}, \quad (11b)$$

$$\mathbf{B}_m^* = F_m \mathbf{V} e^{-i\varphi_m}, \quad (11c)$$

where \mathbf{A} is the free-atom Hamiltonian matrix and \mathbf{V} is the interaction Hamiltonian matrix in the basis set of Eq. (9).

By separating the infinite matrix \mathbf{H}_F in the way shown in Fig. 3 (white and gray shaded areas), the eigenvalue problem is seen to be similar ‘‘topologically’’ to that of the monochromatic case. Hence, the numerical methods for its solution can be carried over from the one color case [1,22,23]. This approach allows the systematic polyelectronic treatment of the polychromatic MEMP problem for any number of commensurate frequencies. However, it is easily seen that the size of the computation and the requirements for computer time increase very rapidly as N_c increases. In this work, we went up to $N_c = 4$, since this is sufficient both as regards the possible execution of experiments and as regards the demonstration of the form that the dependence of the MPIRs has on the phase difference, for weak and for strong fields.

The MEMPT is implemented from first principles by choosing, manipulating, and optimizing appropriate one-electron and N -electron basis sets, representing the electronic

structure and electron correlation of bound, autoionizing, and scattering states. Because of the large size of the problem of diagonalizing \mathbf{H}_F of Fig. 3, it is important to use function spaces which secure that the results contain all the significant physical information, while keeping the computational effort as low as possible. The correlated function space used for the construction of \mathbf{H}_F in this work was the one that was presented in [1]. Therefore, we do not repeat it here. Briefly, the space of ψ_i in Eq. (9) contains bound two-electron functions of real coordinates for the discrete states (ground and Rydberg) up to total angular momentum $L=5$, which are computed by the numerical multiconfigurational Hartree-Fock (MCHF) method [25], state-specific wave functions for the lowest-lying autoionizing doubly excited states, and 82 doubly excited correlating configurations constructed from analytic virtual orbitals, with total $L=0,1,2,3,4$. The space of X_j of Eq. (9) contains \mathcal{L}^2 two-electron functions of the form $1s \otimes \epsilon l$, where the orbital ϵl is expanded in terms of Slater-type orbitals (STO) with a complex coordinate

$$\xi_k(\rho^*) = \xi_k(re^{-i\theta}) = C_k(\theta)r^k e^{-\alpha\rho^*}. \quad (12)$$

For each $l(l=0,\dots,5)$ the continuum orbitals were expanded in ten $\xi_k(\rho^*)$, except for $l=5$, for which eight $\xi_k(\rho^*)$ 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 blocks in \mathbf{H}_F , the complex eigenvalue remains essentially unchanged. Specifically, it was found that convergence was established when the number of the blocks reached ten or eleven, and when the parameters in Eq. (12) are $\alpha=1.5$ and $\theta=0.3$ rad.

IV. RESULTS FOR HE IRRADIATED BY A DICHROMATIC ac FIELD OF $\lambda=248$ nm ($\omega_1=\omega=5$ eV) AND ITS HIGHER HARMONICS ($\omega_2=n\omega$, $n=2,3,4$)

Given the arguments of Sec. II, we divide the results with respect to the order of the harmonics.

A. Even harmonics [$\omega_2=(2k)\omega$]

1. $\omega_2=2\omega$.

Figure 4 shows the MPIR 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/cm² and by its second harmonic with $I_2=6.1 \times 10^{12}$ W/cm², as a function of $\cos(2\varphi_2)$. Even though the MEMPT calculation is nonperturbative, what has emerged is very good linear dependence of the rate on $\cos(2\varphi_2)$, in accordance with the prediction given in Sec. II. The dotted line in Fig. 4 represents the fitted polynomial in powers of $\cos(2\varphi_2)$, to the *ab initio* rates

$$\Gamma = A + B_1 \cos(2\varphi_2) + B_2 \cos^2(2\varphi_2) + \dots, \quad (13)$$

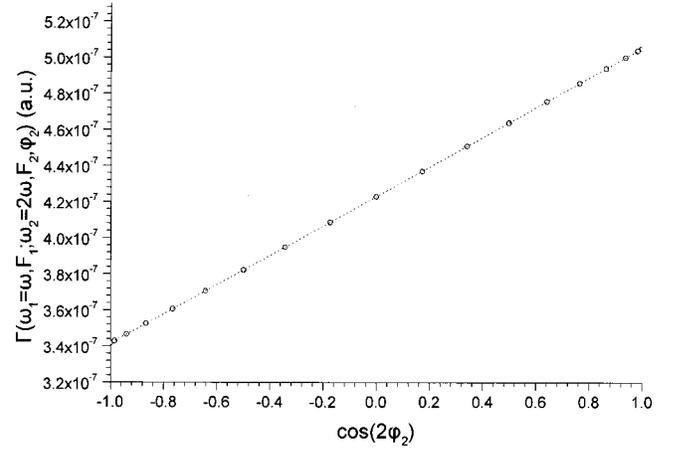


FIG. 4. Multiphoton ionization rates (Γ in a.u.) of the He ground state irradiated by a linearly polarized field of frequency $\omega=0.18373$ a.u. ($\lambda=248$ nm) with intensity $I_1=4.5 \times 10^{13}$ W/cm² and by its second harmonic ($\omega_2=2\omega$) with $I_2=6.1 \times 10^{12}$ W/cm², as a function of $\cos(2\varphi_2)$. The dotted line represents the fitted polynomial of Eq. (13).

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/cm² and $I_2=3.2 \times 10^{13}$ W/cm², the rates deviate from the linear dependence on $\cos(2\varphi_2)$ (Fig. 5). The fitted polynomial of Eq. (13) (dotted line in Fig. 5) yields the following 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 .

2. $\omega_2=4\omega$.

Figure 6 shows the MPIR of He for $I_1=1 \times 10^{14}$ W/cm² and $I_2=1 \times 10^{12}$ W/cm², as a function of $\cos(2\varphi_2)$. 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 one-color MPIR with $\omega=5$ eV, LOPT

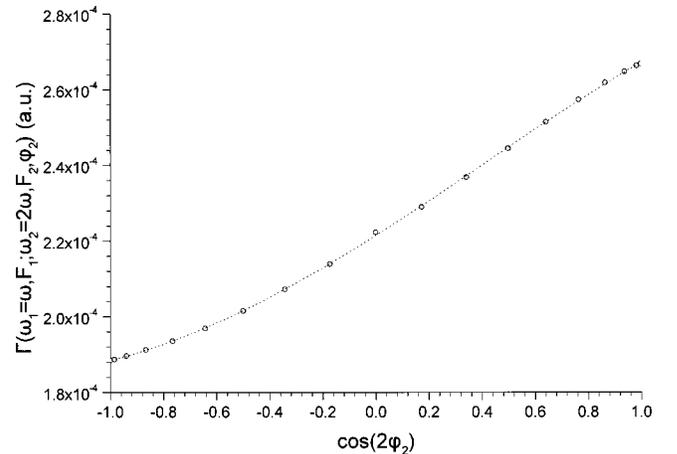


FIG. 5. As in Fig. 4, but for $I_1=3.5 \times 10^{14}$ W/cm² and $I_2=3.2 \times 10^{13}$ W/cm².

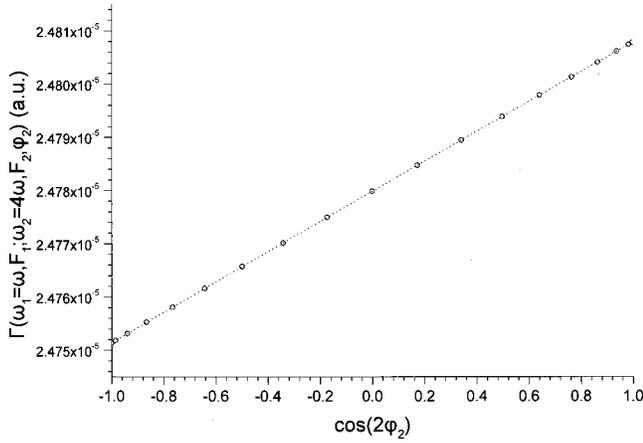


FIG. 6. As in Fig. 4, but for $\omega_2 = 4\omega$, $I_1 = 1 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 1 \times 10^{12} \text{ W/cm}^2$.

breaks down around $I = 7 \times 10^{13} \text{ W/cm}^2$. For weak intensities, five 5-eV photons are needed for ionization of He. However, for $I_1 = 1 \times 10^{14} \text{ W/cm}^2$, six photons are required due to the ponderomotive shift.) The coefficients of the fitted polynomial (dotted line in Fig. 6) are now $A = 2.48 \times 10^{-5} \text{ a.u.}$, $B_1 = 2.83 \times 10^{-8} \text{ a.u.}$, $B_2 = -2.63 \times 10^{-10} \text{ a.u.}$ By increasing the intensities to $I_1 = 2.24 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 3.5 \times 10^{12} \text{ W/cm}^2$, a deviation from the linear dependence is obtained, as is shown in Fig. 7. The values of the coefficients of the fitted polynomial (dotted line) are $A = 1.03 \times 10^{-4} \text{ a.u.}$, $B_1 = 7.62 \times 10^{-8} \text{ a.u.}$, $B_2 = 8.8 \times 10^{-8} \text{ a.u.}$

B. Odd harmonics [$\omega_2 = (2k+1)\omega$]

(1). $\omega_2 = 3\omega$

Figure 8 shows the MPIR of He for $I_1 = 1 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 1 \times 10^{12} \text{ W/cm}^2$, as a function of $\cos(\varphi_2)$. As in Fig. 6, the rate is governed by a linear dependence on $\cos(\varphi_2)$, even though the intensity I_1 is beyond the domain of the LOPT for the monochromatic case. The coefficients of the fitted polynomial (dotted line)

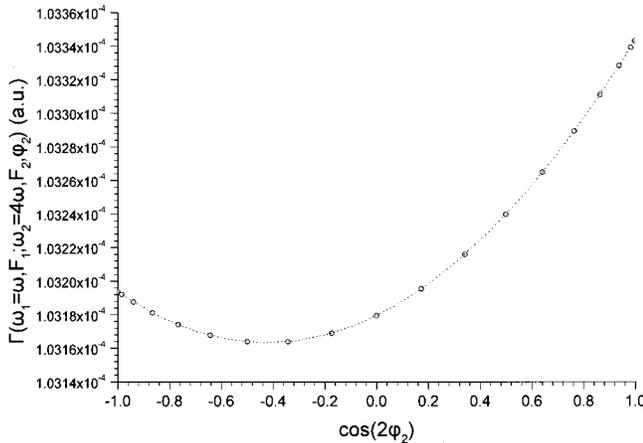


FIG. 7. As in Fig. 6, but for $I_1 = 2.24 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 3.5 \times 10^{12} \text{ W/cm}^2$.

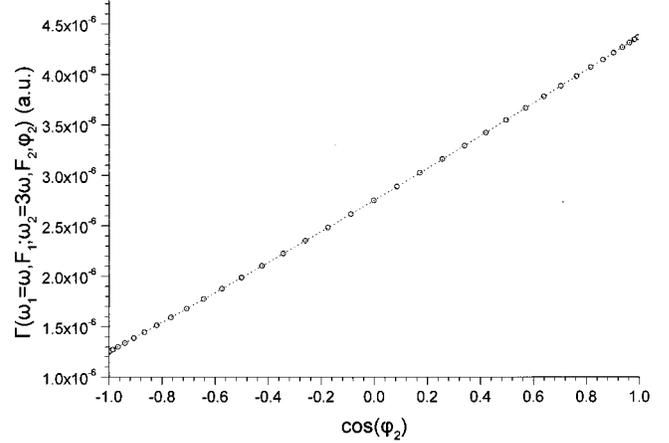


FIG. 8. Multiphoton ionization rates (Γ in a.u.) of the He ground state irradiated by the linearly polarized field of frequency $\omega = 0.18373 \text{ a.u.}$ ($\lambda = 248 \text{ nm}$) with intensity $I_1 = 1 \times 10^{14} \text{ W/cm}^2$ and by its third harmonic ($\omega_2 = 3\omega$) with $I_2 = 1 \times 10^{12} \text{ W/cm}^2$, as a function of $\cos(\varphi_2)$. The dotted line represents the fitted polynomial of Eq. (14).

$$\Gamma = A + B_1 \cos(\varphi_2) + B_2 \cos^2(\varphi_2) + \dots \quad (14)$$

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

An increase of the intensities to $I_1 = 1.7 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 1.4 \times 10^{13} \text{ W/cm}^2$ results in dependence of the rate which is not linear with respect to $\cos(\varphi_2)$. This is shown in Fig. 9. When we fit the polynomial of Eq. (14) to the MEMPT values, the values of the coefficients are $A = 5.04 \times 10^{-5} \text{ a.u.}$, $B_1 = 2.2 \times 10^{-5} \text{ a.u.}$, $B_2 = 3.8 \times 10^{-6} \text{ a.u.}$

V. 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 ($F_1, \omega_1 = \omega; F_2, \omega_2 = m\omega, \varphi_2$), path interference is such that the multiphoton ionization rate is governed by a simple rule as regards its

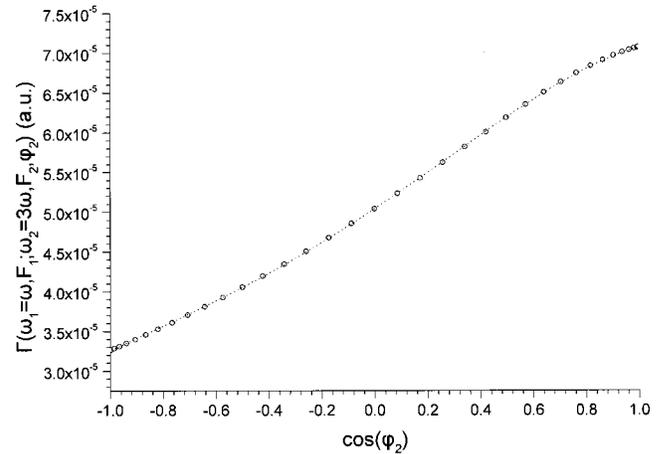


FIG. 9. As in Fig. 8, but for $I_1 = 1.7 \times 10^{14} \text{ W/cm}^2$ and $I_2 = 1.4 \times 10^{13} \text{ W/cm}^2$.

relation to the phase difference φ_2 . Indeed, for odd ($m = 2k + 1$) higher harmonics, the MPIR varies linearly with $\cos(\varphi_2)$, whilst for even higher harmonics it varies linearly with $\cos(2\varphi_2)$. 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 mono-

chromatic case. Given this distinction, wherever the analysis of Sec. II is valid, 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 MPIR on φ_2 is no longer simple, since terms with higher powers of $\cos(\varphi_2)$ and $\cos(2\varphi_2)$ contribute significantly.

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