On the use of the Kramers–Henneberger Hamiltonian in multi-photon ionization calculations

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Abstract
We employ the Kramers–Henneberger Hamiltonian for time-independent calculations of two-photon ionization of atoms with one and two electrons. As compared to the electromagnetic interaction in the length and velocity gauges, the presently employed Kramers–Henneberger gauge has an advantage of the dipole matrix elements for the free–free electron transitions being finite and well-defined quantities. This circumstance simplifies considerably the computations and allows us to obtain accurate results for the two-photon ionization of realistic atomic systems.

1. Introduction

Recent years have witnessed a rapid advancement of high-power short-pulse laser techniques which make it possible to observe many striking phenomena such as multi-photon ionization (MPI) and above-threshold ionization (ATI). This progress in experimental techniques has been accompanied by equally rapid development of theoretical methods needed to describe adequately phenomena occurring in strong laser fields. Representative reviews giving detailed accounts of these theoretical advances can be found in Protopapas et al (1997), Lambropoulos et al (1998), Chu and Telnov (2004) and Posthumus (2004).

It is well known that interaction of an atom and the electromagnetic (EM) field can be described in various ways corresponding to different choices of the gauge. In single-photon ionization calculations, it is the length and velocity gauges that are used most commonly. This choice is quite natural since in the length and velocity gauges the dipole matrix elements assume a very simple form. Direct application of the perturbation theory leads to appearance of formally divergent integrals corresponding to free–free electron transitions. This divergence is in fact artificial due to the neglect of the spatial dependence of the electromagnetic field...
In practical MPI calculations, this circumstance does not pose a major difficulty and several ways have been designed to circumvent it.

For one-electron systems this problem can easily be avoided since analytical expressions for the Coulomb Green function are known. This fact has been exploited in many papers (Klarsfeld 1969, Rapoport et al 1969, Arnous et al 1973, Teague and Lambropoulos 1976, Klarsfeld and Maquet 1979, Karule 1971, 1985, Karule and Moine 2003). Alternatively, one may avoid the divergence problems by reducing summations and integrations over intermediate electron states to solution of nonhomogeneous differential equations (Zernik and Klopfenstein 1965, Chan and Tang 1969, Jayadevan and Thayyullathil 2001).

For systems with more than one electron, where these techniques cannot be implemented, other methods have been developed. The states belonging to the continuous spectrum of the system can be represented by means of a suitable set of the square integrable ($L^2$) functions (Tang and Bachau 1993, Venuti et al 1996). The system is quantized in a box of sufficiently large dimensions which gives a discretized representation of the continuous spectrum. This approach, supplemented by the B-splines technique (Bachau et al 2001), allowed the obtaining of a set of accurate MPI and ATI cross-sections for many-electron systems such as He (Venuti et al 1996, Saenz and Lambropoulos 1999, Nikolopoulos and Lambropoulos 2001) and Be (Tang and Bachau 1993). Alternatively, one may regard the free–free matrix elements as distributions (Veniard and Piraux 1990) rather than ordinary functions and use methods of the distribution theory such as various regularization techniques (Mercouris and Nicolaides 1989, Nicolaides and Mercouris 1989, Korol 1997).

In the present paper we describe an alternative method allowing us to compute amplitudes of the two-photon processes in many-electron systems in a more direct way. The states belonging to the continuous spectrum of the system can be represented by means of a suitable set of the square integrable ($L^2$) functions (Tang and Bachau 1993, Venuti et al 1996). The system is quantized in a box of sufficiently large dimensions which gives a discretized representation of the continuous spectrum. This approach, supplemented by the B-splines technique (Bachau et al 2001), allowed the obtaining of a set of accurate MPI and ATI cross-sections for many-electron systems such as He (Venuti et al 1996, Saenz and Lambropoulos 1999, Nikolopoulos and Lambropoulos 2001) and Be (Tang and Bachau 1993). Alternatively, one may regard the free–free matrix elements as distributions (Veniard and Piraux 1990) rather than ordinary functions and use methods of the distribution theory such as various regularization techniques (Mercouris and Nicolaides 1989, Nicolaides and Mercouris 1989, Korol 1997).

In the present paper we describe an alternative method allowing us to compute amplitudes of the two-photon processes in many-electron systems in a more direct way. The proposed method is based on the so-called Kramers–Henneberger (KH) form of the interaction Hamiltonian (Kramers 1956, Henneberger 1968). The KH representation of the system ‘atom plus electromagnetic field’ is often used when one studies a temporal evolution of atomic systems subjected to a pulse of EM radiation (Reed and Burnett 1991, Vivirito and Knight 1995). The KH representation also enables simple asymptotic boundary conditions used in the external region of the $R$-matrix Floquet theory (Burke et al 1991).

In the present paper we shall be interested in another aspect of the KH representation, namely the advantages its use may offer in the perturbative computations of MPI rates. To our knowledge, the KH description of the EM radiation interaction with atoms has not been used in this context. As we shall see, in the perturbative calculations the KH representation offers one important advantage. In contrast to the length and velocity gauges, in the KH formulation all the dipole matrix elements are finite and well defined. For the laser fields of not very large intensities (below $10^{13}$ W cm$^{-2}$), the perturbation theory (PT) provides quite an adequate description of the MPI process and allows us to achieve numerically accurate results with much less computational labour. We shall consider below two-photon ionization processes in the hydrogen and helium atoms. The highly accurate perturbative results available in the literature allow us to evaluate directly the accuracy of the method. We also discuss some subtle numerical aspects of application of the KH Hamiltonian in perturbative calculations. The use of the KH Hamiltonian in time-independent MPI calculations is not restricted to perturbative regime. In our recent paper (Ivanov and Kheifets 2005) we demonstrated utility of this method in a non-perturbative calculation of the MPI rates in atomic hydrogen.

In the following sections, we briefly recall main theoretical aspects of the KH representation, describe the computation of the dipole matrix elements and apply the technique to two-photon ionization of atomic hydrogen and helium.
2. Theory

A starting point for the description of the interaction of an atom and a monochromatic EM field is the minimal coupling Hamiltonian:

\[ \hat{H} = \hat{H}_\text{atom} + \hat{H}_\text{int}. \]  

(1)

Here \( \hat{H}_\text{atom} \) has the usual meaning of the atomic Hamiltonian:

\[ \hat{H}_\text{atom} = \sum_{i=1}^{N} \frac{p_i^2}{2} - \sum_{i=1}^{N} \frac{Z}{r_i} + \sum_{i,j=1,i>j}^{N} \frac{1}{r_{ij}}. \]  

(2)

The atomic Hamiltonian is taken in a non-relativistic form with \( Z \) being the nucleus charge.

The part of the Hamiltonian \( \hat{H}_\text{int} \) which describes the interaction of the atom and the EM field can be written as (see, e.g., (Sobelman 1972))

\[ \hat{H}_\text{int} = -\frac{1}{c} \sum_{i=1}^{N} \left( \hat{\mathbf{A}} \cdot \hat{p}_i - \frac{\hat{A}^2}{\mathbf{c}^2} \right), \]  

(3)

where \( \hat{p} \) is the momentum operator, \( \hat{\mathbf{A}} \) is a vector potential, summation runs over all atomic electrons. In the following we shall assume that the dipole approximation is valid so that the vector potential does not depend on atomic coordinates. Performing a suitable canonical transformation of the Hamiltonian (3), one can obtain various forms of the interaction Hamiltonian. The KH Hamilton is obtained by the canonical transformation \( \hat{H}_{KH} = e^{i\hat{T}} \hat{H}_\text{min} e^{-i\hat{T}} - \frac{\partial T}{\partial t} \) generated by the operator:

\[ \hat{T} = -\frac{1}{c} \int_{0}^{\tau} \sum_{i=1}^{N} \hat{\mathbf{A}}(\tau) \hat{p}_i \, d\tau + \frac{1}{2c^2} \int_{0}^{\tau} \hat{A}^2(\tau) \, d\tau. \]  

(4)

Expressed in quantum-mechanical terms (as far as description of the EM field is concerned), this transformation is also known as the Pauli–Fierz canonical transformation (Pauli and Fierz 1938). We shall not distinguish between these two versions of the transformation as the final results are identical.

Under transformation (4) the minimal-coupling Hamiltonian (2) becomes

\[ \hat{H}_{KH} = \hat{H}_\text{atom} + \hat{H}_{KH\text{int}}, \]  

(5)

where \( \hat{H}_\text{atom} \) retains the same form as the Hamiltonian (2) while the interaction Hamiltonian becomes

\[ \hat{H}_{KH\text{int}} = \sum_{i=1}^{N} \left( \frac{Z}{r_i} - \frac{Z}{|r_i + \hat{\alpha}|} \right). \]  

(6)

We shall be interested in the case of a linearly polarized monochromatic EM field. In this case \( \hat{\alpha} = \hat{F}/\omega^2 \) where \( \hat{F} \) is the operator of the electric field intensity, \( \omega \) is the photon energy\(^2\). If \( \hat{\alpha} \) can be considered as a small quantity, the leading term of expansion of (6) reproduces the well-known \( Zr/r^3 \) form of the interaction Hamiltonian in the acceleration gauge. This form is often used in the first-order perturbation calculations. We, however, are interested in higher order effects and must generally retain complete form of the Hamiltonian (6).

To build the perturbation theory expansion, treating operator (6) as a perturbation, we need a formula for the matrix elements of this operator sandwiched between the states describing the noninteracting atom and the EM field. It is convenient for our purposes to use the notation

\(^2\) We use the atomic system of units in which \( \hbar = e = m = 1 \).
for these states where \( a \) stands for a set of quantum numbers describing the atom and \( m \) denotes a number of laser photons in a given mode. Such a formula can be obtained from the known matrix elements of the quantized vector potential operator (Ivanov and Kheifets 2005). A simpler derivation, relying on the correspondence between the quantum and classical description of the EM field, is given in appendix A.

Obtained in either way, the final formula reads

\[
\langle a, n + p | \hat{H}_{\text{KH}} | b, n \rangle = \frac{1}{\pi} \sum_{i=1}^{N} \int_{0}^{\pi} \cos p\theta \langle a | \frac{Z}{r_i} - \frac{Z}{|r_i + F \cos \theta/\omega^2|} | b \rangle \, d\theta.
\]

Here \( F \) is already a classical vector and not an operator. Its magnitude is related to the number of photons via the flux conservation relation \( F^2/8\pi = n\omega \), and it is directed along the polarization vector of the incoming photons. In the following, we shall take this direction as the \( z \)-axis.

To amend equation (7) to a form suitable for practical computations, we use the well-known expansion:

\[
\frac{1}{|r + F \cos \theta/\omega^2|} = \sum_{k=0}^{\infty} \sqrt{\frac{4\pi}{2k + 1}} \frac{r_<^k}{r_>^{k+1}} [-\text{sign}(\cos \theta)]^k Y_{k0}(r),
\]

where \( r_< \) (\( r_> \)) is the smaller (greater) of \( r \) and absolute value of \( F \cos \theta/\omega^2 \). Equation (8) allows separation of the radial and angular variables. Angular parts are evaluated analytically using integrals of products of several spherical functions (Varshalovich et al 1988):

\[
\int Y_{l_1 m_1}(\Omega) Y_{l_2 m_2}(\Omega) Y_{l_3 m_3}(\Omega) \, d\Omega = \frac{(2l_1 + 1)(2l_2 + 1)(2l_3 + 1)}{4\pi} \times \left( \begin{array}{ccc} l_1 & l_2 & l_3 \\ 0 & 0 & 0 \end{array} \right) \left( \begin{array}{ccc} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{array} \right).
\]

Equation (9) is written for the case when the atomic subsystem contains one electron (hydrogen). Generalization for the case of many-electron systems is a simple exercise in angular momentum algebra.

We shall be interested in applications of equation (8) in the context of perturbation theory when \( \hat{H}_{\text{KH}} \) can be considered as a perturbation. When performing perturbation calculations, we are usually interested in keeping track of field dependences of the matrix elements. Suppose we study a process for which, in the leading order, the amplitude is proportional to \( k \)th power of electric field strength \( M \propto F^k \), with some integer \( k \). Then we would like to retain in the perturbation theory expressions only the terms which give rise to such dependence in the limit of small \( F \). If the length or velocity forms for the atom–EM field interaction is used, such a count of powers of \( F \) is trivial, following from the well-known selection rules for the matrix elements. In the KH representation, there are no exact selection rules. Nevertheless, the count of powers of the electric field strength is still possible. Consider, for example, the case of a two-photon ionization of a one-electron atom, which is the second-order process. Suppose, we are interested in the ionization from the state \( a \) with a given orbital momentum \( l \). Then it is easy to see from equation (8) that the following asymptotics hold for \( F \to 0 \):

\[
\langle a l m | \hat{H}_{\text{KH}}^{\text{KH}} | b l m \pm 2 \rangle \propto F^2, \quad \langle a l m | \hat{H}_{\text{KH}}^{\text{KH}} | b l \pm 1 m \pm 1 \rangle \propto F.
\]

Here \( a \) and \( b \) stand for the set of all atomic quantum numbers except the angular momentum, the integer \( m \) refers to a total number of laser photons. It is easy to see from equation (8) that the coefficient of proportionality in the second matrix element of (10) is just a matrix element of the operator \( Zr/r^3 \) which is commonly used in the first-order calculations in the acceleration gauge.
Consider the case when the final state $b$ of the atom has the same angular momentum $l$ as the initial state $a$. In the first order of the perturbation theory, the amplitude of the process of two-photon ionization is given by the matrix element:

$$M_1(\alpha l m \rightarrow \beta l m - 2) = \langle \alpha l m | \hat{H}_{\text{KH}}^{\text{int}} | \beta l m - 2 \rangle.$$  

(11)

According to the estimation given above this matrix element $M_1(\alpha l m \rightarrow \beta l m - 2) \propto F^2$, when $F \rightarrow 0$. In the second order of the perturbation theory the corresponding amplitude is given by

$$M_2(\alpha l m \rightarrow \beta l m - 2) = \sum_{c l' = l \pm 1} \frac{\langle \alpha l m | \hat{H}_{\text{KH}}^{\text{int}} | c l' m - 1 \rangle \langle c l' m - 1 | \hat{H}_{\text{KH}}^{\text{int}} | \beta l m - 2 \rangle}{E_{\alpha l} - E_{c l'} + i \epsilon}.$$  

(12)

Here the symbol $\sum$ indicates the sum over the discrete spectrum and integration over continuum of the intermediate states of atomic Hamiltonian. In equation (12) index $l'$ stands for angular momentum of a state belonging to the set of intermediate states, index $c$ refers to all other quantum numbers. From the above estimate for the matrix elements it follows that $M_2(\alpha l m \rightarrow \beta l m - 2) \propto F^2$. The amplitudes of the first and second order are thus of the same order in $F$ and should both be computed. This is, of course, a consequence of the above-mentioned fact that there are no exact selection rules for the matrix elements if we use the Kramers–Henneberger form of the Hamiltonian. If we used length or velocity gauge to describe two-photon ionization, the term $M_1$ would strictly be zero due to the selection rules in these gauges, and we would have to consider only the second-order amplitude $M_2$. In the present approach, as we have seen, we have to retain in the calculation $M_1$ and $M_2$, since, generally, they are both nonzero quantities of the same order in $F$. Terms of the perturbation series of order higher than two will contain at least one additional matrix element of $\hat{H}_{\text{KH}}^{\text{int}}$ and will, therefore, be of the order $F^k$ with $k > 2$. Thus, for the amplitude of the process of two-photon ionization we obtain an expression:

$$M(\alpha l m \rightarrow \beta l m - 2) = M_1(\alpha l m \rightarrow \beta l m - 2) + M_2(\alpha l m \rightarrow \beta l m - 2) + O(F^3),$$

(13)

where the retained terms are of the order of $F^2$. Gauge invariance of the theory guarantees, of course, that the overall coefficient with $F^2$, which we obtain in this way, will be the same (apart from the insignificant phase factors) as in the length or velocity gauges. The previous remark can be used to get a somewhat deeper insight into properties of the perturbation theory one obtains treating the Hamiltonian $\hat{H}_{\text{KH}}^{\text{int}}$ as a perturbation. Applying standard prescriptions of the perturbation theory, we can write a formal series in ‘powers’ of $\hat{H}_{\text{KH}}^{\text{int}}$ for an amplitude of any given process. Terms of this formal series are expressed via the matrix elements of $\hat{H}_{\text{KH}}^{\text{int}}$ and can be expanded in powers of electric field strength $F$ by means of equation (8). Substituting these expansions into the perturbation series, we can rewrite perturbation series for the amplitude as an expansion in powers of $F$. Gauge invariance guarantees, that retaining finite number $N$ of terms of this series, will give for any physical quantity the same result, we would obtain if we performed perturbative calculation (of the same order $N$) using the length or velocity gauges. In, particular, this means that the contribution of all terms of the order higher than $F^2$ in equation (13) will be the same as in the length or velocity gauges. The convergence properties of the series (13) are, therefore, essentially the same as those of analogous expressions in the length or velocity gauges. The remainder of the series and, hence, the error which neglect of terms of the higher order introduces will be the same as in other gauges.
Equation (13) was written for the final atomic state $bl$ having the same angular momentum as the initial state. It is easy to see that the same reasoning applies to other possible final channels $bl + 2, bl - 2$ of the process of two-photon ionization.

Summarizing all said above, the leading term of the amplitude of two-photon ionization is a sum of the matrix elements of the first and second order, which are both of the same magnitude ($\propto F^2$). For various final channels the corresponding formulae are

$$M(al m \rightarrow bl m - 2) = \langle al m|\hat{H}_{\text{int}}^{\text{KH}}|bl m - 2\rangle$$

$$+ \sum_{cl'm = l+1} \frac{\langle al m|\hat{H}_{\text{int}}^{\text{KH}}|cl'm - 1\rangle\langle cl'm - 1|\hat{H}_{\text{int}}^{\text{KH}}|bl m - 2\rangle}{E_{al} - E_{cl'} + i\epsilon}$$

(14)

$$M(al m \rightarrow bl + 2 m - 2) = \langle al m|\hat{H}_{\text{int}}^{\text{KH}}|bl + 2 m - 2\rangle$$

$$+ \sum_{cl'm = l+1} \frac{\langle al m|\hat{H}_{\text{int}}^{\text{KH}}|cl'm - 1\rangle\langle cl'm - 1|\hat{H}_{\text{int}}^{\text{KH}}|bl + 2 m - 2\rangle}{E_{al} - E_{cl'} + i\epsilon}$$

(15)

$$M(al m \rightarrow bl - 2 m - 2) = \langle al m|\hat{H}_{\text{int}}^{\text{KH}}|bl - 2 m - 2\rangle$$

$$+ \sum_{cl'm = l-1} \frac{\langle al m|\hat{H}_{\text{int}}^{\text{KH}}|cl'm - 1\rangle\langle cl'm - 1|\hat{H}_{\text{int}}^{\text{KH}}|bl - 2 m - 2\rangle}{E_{al} - E_{cl'} + i\epsilon}$$

(16)

Without sacrifice of accuracy, in the spectral sums we may use the operator $Z r^3$ instead of the complete form of the operator $\hat{H}_{\text{int}}^{\text{KH}}$.

Once amplitudes (14)–(16) are computed, the generalized partial cross-section of the two-photon ionization from the initial state $a, l$ to a final channel $b, l'$ is given by (cf (Tang and Bachau 1993))

$$\sigma(al \rightarrow bl') = 27\pi^3\alpha^2 a_0^4\tau_0\omega^2 \lim_{F \rightarrow 0} \frac{|M(al \rightarrow bl')|^2}{F^4 k} (\text{cm}^4 \text{s}^{-1}).$$

(17)

Here $\alpha, a_0, \tau_0$ are the fine structure constant, the atomic unit length in centimetres and the atomic unit time in seconds. $F$ is the EM field strength and $\omega$ is the photon energy, both expressed in the atomic units. The one-electron continuum wavefunctions used to calculate the ionization amplitudes are normalized on the momentum scale. The generalized cross-section (17) is related to the ionization rate

$$\Gamma(al \rightarrow bl') = \frac{\sigma(al \rightarrow bl') \times 10^{12}\lambda\text{Ryd}}{13.605 \times 1.602 19} (\text{W}^{-1}\text{cm}^4),$$

where $\lambda$ is the wavelength (in nm) and Ryd $= 109 677$ cm$^{-1}$ is the Rydberg constant (Karule and Moine 2003).

It has to be noted that the KH transformation modifies, in general, the atomic states after the field is switched off at a sufficiently large time (Vivirito and Knight 1995, Reed and Burnett 1991). However, it is shown in appendix B that this does not affect the cross-sections or ionization rates and can only be noted in the fine details of the temporal evolution of the system.

3. Numerical computations

A certain amount of care has to be exercised when amplitudes (14)–(16) are computed numerically. Consider matrix elements in the sums over intermediate states in these expressions. Separating radial and angular variables with the help of equations (7) and (8), and taking the form $Z r^3$ for the interaction Hamiltonian we obtain integrals of the sort

$I = \int R_{el}(r)R_{el'}(r) dr$. Here $R$ are radial electron wavefunctions behaving as $r^l$ near the
origin \( r = 0 \). Function \( R_{EI} \) describes either an initial or final state of the process, \( R_{EI} \) is the radial wavefunction of the intermediate state belonging to the continuous spectrum which, for large momenta, behaves as \( R_{EI}(r) \propto \sin(kr + \delta)/r \). The \( k \)-dependence of the integral is crucially dependent on the orbital momentum \( l \). If \( l > 0 \), the integral \( I \) can be approximated for large \( k \) as \( \int R_{EI}(r) \sin(kr + \delta)/r \) (we omit unimportant normalization factors). Consider the function \( P(r) = R_{EI}/r \). If \( l > 0 \), \( \int_0^{\infty} |P(r)| \, dr \) is finite, hence the Riemann–Lebesgue lemma is applicable and the integral \( I \) will decay for \( k \to \infty \). If \( l = 0 \), the replacement of the \( R_{EI} \) by its asymptotic expression is not legitimate (we would obtain a divergent integral). More careful study shows that if \( l = 0 \) then \( I \to \) const when \( k \to \infty \). The integrals over momenta of intermediate states in formulae (14)–(16) still converge in this case due to the energy denominator but much more slowly than in the case of \( l > 0 \). This means that to achieve a good numerical accuracy for the amplitudes with \( l = 0 \), one must take into account an asymptotic tail of the integrand for \( k \to \infty \).

In practice, this does not pose serious difficulties. One has only to determine (either analytically or numerically) the constant in the expression \( I \to \) const for \( k \) large enough so that this asymptotic law holds well, and then add the corresponding contribution to the integrals in formulae (14)–(16). For \( l > 0 \) one need not worry about the asymptotic tails of the integrand as integrals \( I \) decay quite fast with \( k \).

For the presently considered targets (H and He), the initial state has an S orbital character leading to the two final channels S and D which correspond to amplitudes (14) and (15). As explained above, we may retain in these amplitudes only the \( P \) intermediate states. Both the discrete and continuous intermediate states were taken into account. The continuous spectrum integration follows closely prescriptions given in Bray (1994) and Ivanov and Kheifets (2005). The interval of momenta \((0, q_{\text{max}})\) is divided into several subintervals. For the photon energies above the ionization threshold, a pole is present in the momentum integral. To carry out the integration around the pole accurately, the first two subintervals are chosen to be \((0, k_{\text{pole}})\) and \((k_{\text{pole}}, 2k_{\text{pole}})\) with a typical number of 20 momentum points in each subinterval. Then the delta-function singularity is isolated and the remaining principle value integral is evaluated by a modified Gaussian rule (Bray 1994). The remaining part of the momentum integral is divided as follows: \((2k_{\text{pole}}, 4)\) (20 integration points), \((4, 10)\) (20 points) and \((10, q_{\text{max}})\) (20 points). These intervals are pole-free and the integration is performed by using a Gauss quadrature rule. The fairly large value of \( q_{\text{max}} \) is chosen to take care of a slow decay of the integrand in the \( S \)-channel. The asymptotic tail \((q_{\text{max}}, \infty)\) is calculated analytically.

Once the amplitudes are computed the partial cross-sections and rates can be determined via equations (17) and (18). Total cross-sections and rates corresponding to linear and circular polarization of the EM field can then be determined. If the initial state of the atomic system is an S-state, then: \( \Gamma^\pm(ns) = \Gamma(ns \to ks) + \Gamma(ns \to kd) \) (linear polarization) and \( \Gamma^\circ(ns) = \frac{1}{2} \Gamma(ns \to kd) \) (circular polarization) (Jayadevan and Thayyullathil 2001).

All calculations reported below were performed for the EM field strength of \( F = 0.03 \) au. This field strength is small enough, so that determination of the cross-sections via equation (17) for small but finite \( F \) will be, as we shall see, quite accurate.

4. Results

4.1. Hydrogen

In the case of hydrogen, all the one-electron wavefunctions are known analytically. These allowed us to include 15 discrete intermediate \( P \) states and carry out integration up to \( q_{\text{max}} = 70 \) au in the second-order amplitudes.
Table 1. Ionization rates (in units of W$^{-1}$ cm$^4$) for the two-photon ionization of atomic hydrogen in the ground state by linearly $\Gamma^l$ and circularly $\Gamma^c$ polarized light. Numbers in parentheses indicate powers of 10. The literature values are denoted as JT01 (Jayadevan and Thayyullathil 2001) and KM03 (Karule and Moine 2003).

| $\lambda$ (nm) | $\Gamma^l$ JT01 | $\Gamma^l$ KM03 | $\Gamma^l$ Present | $\Gamma^c$ JT01 | $\Gamma^c$ KM03 | $\Gamma^c$ Present |
|---------------|----------------|----------------|-------------------|----------------|----------------|-------------------|
| 20            | $3.02(-38)$    | $3.01(-38)$    | $3.02(-38)$       | $2.44(-38)$    | $2.43(-38)$    | $2.44(-38)$       |
| 40            | $2.15(-36)$    | $2.14(-36)$    | $2.15(-36)$       | $2.03(-36)$    | $2.02(-36)$    | $2.03(-36)$       |
| 60            | $2.62(-35)$    | $2.61(-35)$    | $2.62(-35)$       | $2.79(-35)$    | $2.78(-35)$    | $2.79(-35)$       |
| 80            | $1.58(-34)$    | $1.57(-34)$    | $1.59(-34)$       | $1.85(-34)$    | $1.84(-34)$    | $1.86(-34)$       |

In table 1, we present the rates of two-photon ionization of the ground state hydrogen atom by linear and circular polarized light and compare them with the latest literature values (Jayadevan and Thayyullathil 2001, Karule and Moine 2003). The three sets of data are virtually identical for all the photon energies considered. The cited literature values have been obtained analytically which is possible for hydrogen. They can be, therefore, considered as ‘exact’. The comparison in the table indicates that the use of the KH Hamiltonian allowed us to achieve comparable level of accuracy with little effort.

4.2. Helium

We consider now two-photon ionization from the ground state of helium. Due to possible two-electron excitations, complete treatment of this problem is much more complicated than for hydrogen. However, for the photon energies below the threshold of the $N = 2$ excitations, we can use a frozen core approximation and restrict ourselves with only one active electron. In this approximation, the problem is effectively reduced to a hydrogen-like calculation with numerical Hartree–Fock wavefunctions.

We describe the helium atom as follows. For the ground state we use the self-consistent Hartree–Fock approximation (Chernysheva et al 1976). The ground state is thus represented as a product of the two 1s orbitals. For the excited states, both discrete and continuous, we use the frozen core Hartree–Fock approximation (Chernysheva et al 1979). These states are thus represented as properly symmetrized products of the 1s core orbital and an orbital describing excited electron either in the bound state or the continuum.

Because of this frozen-core approximation, the calculation for helium proceeds in almost exactly the same way as for hydrogen. The only difference is that due to the equivalence of the 1s$^2$ electrons an additional factor of 2 arises in the formula for the cross-section. As for hydrogen, we may retain only P states in the sums over the intermediate states. We have retained seven P states corresponding to the excitations 1s$n$p with $n = 2–8$. Integration over the continuous spectrum was performed as for hydrogen except for the value of $q_{\text{max}} = 30$ au. All we said above concerning importance of the correct account of the asymptotic tail in the momentum integral for the S-wave, applies for the case of helium as well.

Our results are presented in figure 1 in comparison with other calculations (Saenz and Lambropoulos 1999, Nikolopoulos and Lambropoulos 2001) which use a considerably more accurate representation of the helium atom. Despite a rather crude character of the frozen-core approximation, in the region of the photon energies considered we achieve quite a satisfactory agreement with the literature values. We could not, of course, extend our calculation into the region of larger photon energies since there the processes of core-excitations become essential.
5. Conclusion

We have shown that the KH description of the atom–EM radiation interaction can be used efficiently in calculations of two-photon processes in realistic atomic systems. The fact that the dipole matrix elements between continuous electron states are finite and well-defined quantities makes the calculation relatively simple, both numerically and conceptually.

For hydrogen, the present results agree completely with those obtained in Jayadevan and Thayyullathil (2001) and Karule and Moine (2003). As the latter results are analytical and can therefore be considered as virtually exact, we may be confident that the present approach allows one to achieve quite a high accuracy. Our method can also be applied to the systems with more than one electron, as the comparison with the data for the two-photon ionization of helium shows. Despite the fact that we used rather a crude description of the field-free helium atom (we omitted the core-excitation effects), we obtained good quantitative agreement with the results of other authors who employed a more elaborate representation for the helium atom. As was indicated above, the accuracy of our description of the process of two-photon ionization of helium can further be improved without any problems of conceptual character. All we have to do is to ‘thaw’ the core and to allow the core excitations. This can be done, for example, with the use of the convergent close-coupling (CCC) method (Bray 1994) which is known to provide good description of a complete set of two-electron states, both discrete and continuous. Such calculation will be reported elsewhere.

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Appendix A

We give below a derivation of equation (7) for the matrix elements of the KH interaction Hamiltonian based on the well-known correspondence between the classical Floquet and the quantum-mechanical descriptions of the atom–EM field interaction (Shirley 1965).
In the classical picture, the KH Hamiltonian has the form (6) with \( \alpha = F \cos \omega t/\omega^2 \) where \( F \) is a classical amplitude of the EM field. With this expression being a periodic function of time, the Schrödinger equation has a set of solutions (the Floquet anzats) which allows the following Floquet–Fourier expansion:

\[
\Psi(t) = e^{-iE t} \sum_{n=-\infty}^{\infty} u_n e^{in\omega t},
\]

where \( E \) is the quasi-energy. Expanding the time-periodic function \( \hat{H}_{\text{int}} \) as a Fourier series,

\[
\hat{H}_{\text{int}}^{\text{KH}}(t) = \sum_{n=0}^{\infty} V_n \cos n\omega t,
\]

with

\[
V_n = \frac{2\omega}{\pi} \int_0^{\pi/\omega} \hat{H}_{\text{int}}^{\text{KH}}(wt) \cos n\omega t \, dt,
\]

and equating coefficients with \( e^{-im\omega t} \), one obtains a set of equations for the Fourier amplitudes \( u_n \):

\[
(E - n\omega - \hat{H}_{\text{atom}}) u_n = \sum_{\substack{i,m, j \neq 0 \atop n - i = m}} V_i \frac{1}{2} u_m + \sum_{\substack{i,m, j \neq 0 \atop n + i = m}} V_i \frac{1}{2} u_m.
\]

In the quantum-mechanical description, the coefficients with the amplitudes \( u_m \) on the rhs of equation (A.4) are nothing but the matrix elements \( \langle n | \hat{H}_{\text{int}}^{\text{KH}} | m \rangle \) taken between the states with \( n \) and \( m \) photons (Shirley 1965). This correspondence holds if we neglect all spontaneous processes and retain only laser photons. Since in equation (A.4) summation index \( k \geq 0 \), for given \( n, m (n \neq m) \), the two terms on the rhs of this equation can be combined to give rise to \( V_{|n-m|} \). This gives immediately formula (7) for the matrix elements of the operator \( \hat{H}_{\text{int}}^{\text{KH}} \). In our earlier paper (Ivanov and Kheifets 2005), we obtained this formula directly using quantized form of the electric field operator.

Appendix B

A comment has to be made on the applicability in the present case of the Fermi golden rule which is used to derive equation (17). Below, all the discussion uses only classical terms for the description of the EM field which leads to the same results as the full quantum-mechanical treatment but is somewhat simpler.

Physically, transformation to the KH frame is equivalent to transformation to the non-inertial frame oscillating with the electron. Let us suppose that an initially field-free atom is in some state \( \Psi_0 \). At the moment \( t = 0 \) the interaction of the atom and EM field is switched on and it is switched off at the moment \( t = t_1 \) with some \( t_1 \) large enough so that all the transient processes are negligible. With the help of formulae (14)–(16) we can obtain (for sufficiently large \( t_1 \)) perturbative solution of the time-dependent Schrödinger equation in the KH-frame. As a result, we obtain at the moment \( t_1 \) a vector in the KH frame:

\[
\Psi_{\text{KH}}(t_1) = U^{\text{KH}}(0, t_1) \Psi_0,
\]

where \( U^{\text{KH}}(0, t_1) \) is the evolution operator which we constructed in the KH frame to a given order of the perturbation theory.

To determine rates of various processes, we must now find overlaps of the vector thus obtained with various states of the field-free atom. The latter, however, are generally different
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from the eigenstates of $H_0$, the original field-free atomic Hamiltonian. As formula (4) shows, they are connected to the eigenstates of $H_0$ by means of a unitary transformation generated by the operator $\hat{T}$ in equation (4). The fact that final states are generally altered by this operator may play an important role in calculations of time-evolution of an atomic system under the action of an impulse of EM radiation (Vivirito and Knight 1995, Reed and Burnett 1991).

The problem we are considering here differs from the problems of determining the time-evolution under the action of an impulse of EM radiation in one important aspect. We are at liberty to switch off interaction at any time (provided it is large enough). Operator $\hat{T}$ in equation (4) contains two terms. The one proportional to the square of the vector potential is unimportant (it is a pure phase factor in the dipole approximation which we use). It is the term linear in $A$ that is responsible for the alteration of the final states. We may use, however, periodicity of the vector potential and choose the moment $t_1$ so that this term disappears. The final states then remain unaltered and the validity of the Fermi golden rule is restored.

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