Lippmann-Schwinger description of multiphoton ionization

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Abstract. We develop a formalism and a computational procedure to treat the process of multiphoton ionization (MPI) of atomic targets in strong laser fields. We treat the MPI process nonperturbatively as a scattering phenomenon by solving a set of coupled integral equations. As the basic building blocks of the theory we use a complete set of field-free atomic states, discrete and continuous. This approach should be able to provide both the total and differential cross-sections of MPI of atoms with one or two electrons. As a test, we apply the proposed procedure to a simple model of MPI from the square well potential.

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1. Introduction

In recent years, the process of multiphoton ionization (MPI) of atomic and molecular species has been a subject of intensive experimental and theoretical studies (see reviews by Protopapas *et al* (1997), Lambropoulos *et al* (1998), Chu and Telnov (2004) and Posthumus (2004) and references therein). Rapid progress in this field has been largely driven by advancement of high-power and short-pulse laser techniques. The laser intensities which may go beyond 10^{13} Wcm^{-2} make it possible to observe many striking phenomena such as MPI or above-threshold ionization.

Accurate theoretical description of ionization processes occurring in laser fields of such intensities is bound to go beyond a simple perturbative picture. The first nonperturbative theory of MPI was proposed by Keldysh (1964), Faisal (1973) and Reiss (1980). Their theory (known as KFR) treated the process of MPI as a transition of an electron from an initial bound state into a final state described by the classical Volkov wave function. The KFR approach provided simple analytical formulas for the MPI rate which were found in a qualitative agreement with experiment. Various modifications of the KFR theory were made, in particular those accounting for the rescattering process (Becker *et al* 1994, Bao *et al* 1996).

The KFR theory treated the laser field purely classically. In the following years the MPI problem was reformulated in an entirely quantum form. The first step in this direction was taken by Guo and Aberg (1988) and Guo, Aberg and Crasemann (1989). Their MPI theory (referred hereafter as GAC) was treating the photoionization process as a QED scattering phenomenon. The emphasis in this theory was put on a proper QED description of an electron interacting with the laser field (the quantum version of the Volkov states). Further development of the QED picture of the MPI phenomenon was made by Gao *et al* (2000) and Chen *et al* (2003) who refined the original GAC theory by including the non-laser modes of the electromagnetic field. This approach, although solving the problem in principle, was found to be somewhat difficult to implement even for simplest atomic targets such as one- and two-electron atoms. For instance, Chen *et al* (2003) had to make further approximations in order to carry out their calculation

of MPI on atomic hydrogen.

In the present paper we propose a scattering formalism for MPI which we intend to use for practical computations on real atomic systems. In this development we are inspired by a series of works by Burke and collaborators who combined the the Floquet description of the laser field with the R-matrix scattering theory. Following the seminal work (Burke *et al* 1991), this approach has been successfully implemented for calculating the total MPI rate and the level shift in atomic hydrogen (Dorr *et al* 1992), helium (Purvis *et al* 1993), the negative hydrogen ion (Dorr *et al* 1995) and molecular hydrogen (Colgan *et al* 2001). Most recently the R-matrix Floquet theory was combined with the basis spline technique to describe the two-electron MPI from the helium atom in the ground (Feng and van der Hart 2003) and excited states (van der Hart and Feng 2001). In addition to the total MPI rate, some differential cross-sections can be also calculated within the Floquet formalism as was demonstrated by Potvliege and Shakeshaft (1988) and Potvliege (1998) for atomic hydrogen. It is the most detailed fully differential cross-sections that are of particular interest to experimentalists and that we intend to calculate in our approach.

It should be noted that the Floquet theories mentioned above treat the MPI process as a decay problem. On the other hand, the scattering MPI theories (Guo and Aberg 1988, Guo *et al* 1989, Chen *et al* 2003, Gao *et al* 2000) use the Volkov states to describe intermediate states of the target between absorption of the photons. This makes it difficult to apply these theories to non-trivial atomic systems, e.g. the helium atom. In the present paper, as building blocks of the collision theory, we use atomic field-free states including those states with one and two electrons. For two-electron atomic systems, an accurate set of target states can be generated by the so-called convergent close coupling (CCC) method. This method has been extensively tested for processes with two electrons in the continuum such as electron scattering on atomic hydrogen (Bray 1994) and low-field double ionization of helium (Kheifets and Bray 1998*b*, Kheifets and Bray 1998*a*). We intend to use the same set of target states for MPI of He in the non-perturbative strong-field regime.

The rest of the paper is organized as follows. In Section 2 we give a formulation of the MPI theory in terms of the field-free atomic states, treating MPI as a scattering phenomenon. In Section 3 we make a connection of the present formalism with the Floquet theory. In Section 4 we consider a model square-well problem. We conclude in Section 5 by outlining a set of problems we intend to consider in the future.

2. MPI as a scattering process.

Let us consider a system consisting of a number of photons (with a given frequency ω and momentum vector $\boldsymbol{k}$, corresponding to those of an incident plane-wave), and a target (atom or ion). We shall describe the field fully quantum-mechanically. The Hamiltonian of the system can be written as:

$$\hat{H} = \hat{H}_{\rm atom} + \hat{H}_{\rm field} + \hat{H}_{\rm int}, \tag{1}$$

where $\hat{H}_{\rm atom}$ and $\hat{H}_{\rm field}$ have usual meaning of the Hamiltonians of atom and free field. The atomic Hamiltonian is taken in a non-relativistic form. The corresponding states of the system consisting of the non-interacting atom and field are denoted as $|a, n\rangle$, where a set of quantum numbers a defines the state of the atom and n is the number of photons. The following notations will be kept throughout the paper: Greek letters will be used to designate the states of a whole system "the atom plus external field", while the Latin letters will be used for the atomic states. The atomic system of units is in use with $e = m = \hbar = 1$.

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

$$\hat{H}_{\rm int} = -\frac{1}{c}\hat{\boldsymbol{A}} \cdot \hat{\boldsymbol{p}} + \frac{\hat{\boldsymbol{A}}^2}{2c^2}, \tag{2}$$

where $\hat{\boldsymbol{A}}$ is a quantized vector potential and $\hat{\boldsymbol{p}}$ is the momentum operator. We restrict ourselves with the dipole approximation in which the operator $\hat{\boldsymbol{A}}$ does not act on the atomic coordinates. We will also ignore all the processes of spontaneous emission and absorption of photons. Therefore, here and below, $\hat{\boldsymbol{A}}$ describes the laser photons only. This approach is justified as we are interested in processes induced by strong fields.

The matrix elements of the vector potential operator taken between the noninteracting states of the system "atom plus laser photons" are given by the well-known formulas (see e.g. Sobelman (1972)):

$$\langle a,n|\hat{\boldsymbol{A}}\cdot\hat{\boldsymbol{p}}|b,n-1\rangle = \sqrt{\frac{2\pi c^2 n}{\omega}}\langle a|\boldsymbol{e}\cdot\hat{\boldsymbol{p}}|b\rangle$$
$$\langle a,n|\hat{\boldsymbol{A}}\cdot\hat{\boldsymbol{p}}|b,n+1\rangle = \sqrt{\frac{2\pi (n+1) c^2}{\omega}}\langle a|\boldsymbol{e}\cdot\hat{\boldsymbol{p}}|b\rangle \qquad (3)$$

where $\boldsymbol{e}$ is the polarization vector. In strong fields $n \simeq n-1 \gg 1$ and the coefficients in Equation (3) can be simplified to

$$\sqrt{\frac{2\pi n c^2}{\omega}} \approx \sqrt{\frac{2\pi (n+1) c^2}{\omega}} \approx \frac{Fc}{2\omega}, \qquad (4)$$

where F is the electric field strength related to the energy density as $F^2 = 8\pi n\omega$. This leads to the following formulas for the matrix elements of the operator $\hat{H}_{\rm int}$:

$$\langle a,n|\hat{H}_{\rm int}|b,n\pm 1\rangle = -\frac{F}{2\omega}\langle a|\boldsymbol{e}\cdot\hat{\boldsymbol{p}}|b\rangle, \qquad (5)$$
$$\langle a,n|\hat{H}_{\rm int}|b,n\pm 2\rangle = \frac{F^2}{8\omega^2}\langle a|b\rangle$$
$$\langle a,n|\hat{H}_{\rm int}|b,n\rangle = \frac{F^2}{4\omega^2}\langle a|b\rangle$$

The two bottom matrix elements can be eliminated by using an orthogonal set of the atomic states such as $\langle a|b\rangle = 0$.

The evolution of the system "the atom plus external field" is described by the scattering T- matrix taken in the post-form

$$T^{\beta,\alpha} = \langle\beta|\hat{H}_{\rm int}|\Psi_\alpha^+\rangle, \qquad (6)$$

where $\alpha \equiv a,n$ and $\beta \equiv b,n\pm 1$ are combinations of the atom and photon variables. The bra-vector $|\beta\rangle$ is a free state of the non-interacting atom and field described by the Hamiltonian $\hat{H}_0 = \hat{H}_{\rm atom} + \hat{H}_{\rm field}$. An exact eigenstate of the system Ψ_α^+ is reduced in the distant past, by way of forming appropriate wave packets, to a physical situation when n photons strike an atom in an initial bound state a.

When the interaction is turned on, the initial state of the system α becomes autoionizing as it can couple with the continuum state of the atom accompanied by a lesser number of photons. A scattering formalism which describes such autoionizing

states has long been known since the work of Fano (1961). Here we adopt this formalism in full. We seek a solution of the Scr"Odinger equation

$$(\hat{H} - E_\alpha)\Psi_\alpha^+ = 0 \tag{7}$$

in the form

$$\Psi_\alpha^+ = c_\alpha|\alpha\rangle + \sum_{\gamma\neq\alpha} c_\gamma|\gamma\rangle, \tag{8}$$

where summation runs through all atomic states and laser modes and $|\alpha\rangle \equiv |a,n\rangle$ represents the initial state of the system "atomic bound state plus n laser photons". Substituting this expansion into (7) and determining the coefficients in a usual way as to satisfy the proper boundary conditions, one arrives at the following equation for the wave function Ψ_α^+

$$\Psi_\alpha^+ = |\alpha\rangle + \sum\!\!\!\!\!\!\int_{\gamma\neq\alpha} \frac{\langle\gamma|\hat{H}_{\rm int}|\Psi_\alpha^+\rangle}{E_\alpha - E_\gamma + i\epsilon}|\gamma\rangle \tag{9}$$

From Equation (9) one can obtain a set of Lippmann-Schwinger equations for the T-matrix describing the interaction of the atom with the photon field which reads:

$$T^{\beta\alpha} = \hat{H}_{\rm int}^{\beta\alpha} + \sum\!\!\!\!\!\!\int_{\gamma\neq\alpha} \frac{\hat{H}_{\rm int}^{\beta\gamma}\, T^{\gamma\alpha}}{E_\alpha - E_\gamma + i\epsilon} \tag{10}$$

Here the sign of $i\epsilon$ gives the rule of bypassing the pole when performing the integration over the continuum spectrum.

In Figure 1 we give a graphical representation of Equation (10). Here a straight line with an arrow to the right represents an electron and the dashed lines are used to depict the photons. A vertex indicated by the dot with two electron lines and one photon line represents a dipole matrix element (6) which incorporates many-electron correlation in the target. A rectangular block stands for the T-matrix (10). In the low-field regime the integral term in the right-hand side of Equation (10) can be ignored and the atomic ionization is described by the bare matrix element $\langle a,n|\hat{H}_{\rm int}|b,n-1\rangle$. The strong field effects are incorporated in the integral term and include multiple absorption and emission of the laser photons.

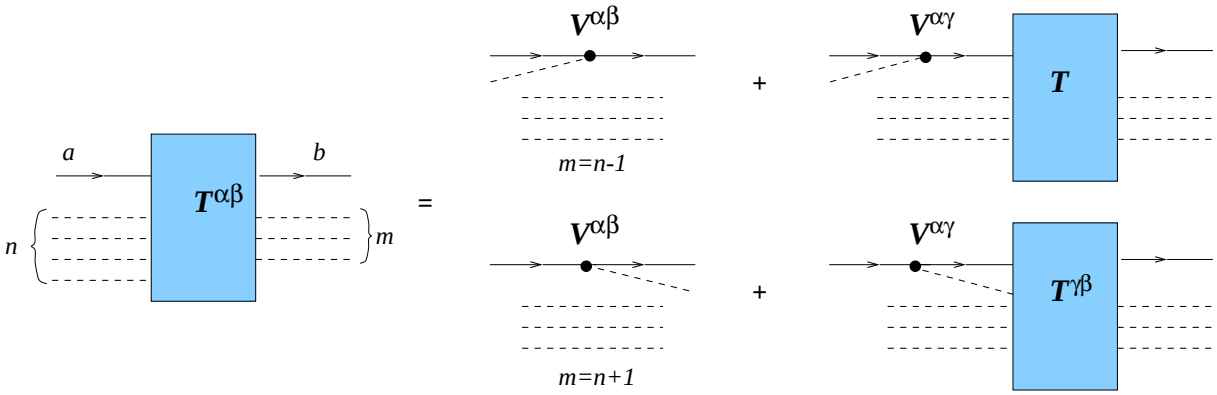

Figure 1. Diagram representation of the Lippmann-Schwinger equation for multiphoton ionization. The graphical symbols are described in the text.

The sum over the spectrum of $\hat{H}_0$ in Equation (10) includes summation over various number of photons as well as summation over bound atomic states and integration over the continuous spectrum of the atom. To compute the integral in (10) a suitable quadrature rule must be chosen by introducing a discrete set of target pseudostates. As the result of such a discretization the set of equations (10) becomes a linear system on the unknown elements of the T-matrix. Once this linear system is solved, all information about the integral and differential features of the MPI process can be obtained from the on-shell elements of the T-matrix. We note that in the weak field limit summation in Equation (10) can be restricted to the atomic variables and we arrive to the expression for the dipole matrix element given by Eq. (12) of Kheifets and Bray (1998*b*).

3. Connection with Floquet formalism

It is instructive to make a connection between the present formalism and that based on the Floquet ansatz (Burke *et al* 1991, Dorr *et al* 1992, Dorr *et al* 1995). To make connections with these works we first apply a unitary transformation:

$$\tilde{\Psi}_\alpha^+ = \exp\left\{-\frac{i}{c}\hat{\boldsymbol{A}}\hat{\boldsymbol{r}}\right\}\Psi_\alpha^+ \tag{11}$$

thus introducing the electromagnetic interaction in the lenght gauge. In this representation the interaction of the electromagnetic field and atomic subsystem assumes the form

$$\hat{H}_{\rm int} = \hat{\boldsymbol{F}}\cdot\hat{\boldsymbol{r}}, \tag{12}$$

with the matrix elements

$$\langle a,n|\hat{\boldsymbol{F}}\cdot\hat{\boldsymbol{r}}|b,n-1\rangle = \sqrt{2\pi n\omega}\langle a|\boldsymbol{e}\cdot\hat{\boldsymbol{r}}|b\rangle$$
$$\langle a,n|\hat{\boldsymbol{F}}\cdot\hat{\boldsymbol{r}}|b,n+1\rangle = \sqrt{2\pi(n+1)\omega}\langle a|\boldsymbol{e}\cdot\hat{\boldsymbol{r}}|b\rangle \qquad (13)$$

The resulting wave function of the system "the atom plus external field" is expanded over a complete set of the free photon states $|\nu\rangle$:

$$\tilde{\Psi}_\alpha^+ = \int u_\nu(\tau)\ |\nu\rangle\ d\nu\ . \qquad (14)$$

Here ν stands for a totality of all quantum numbers defining a state of the photonic system and τ denotes the set of variables describing the atomic subsystem. We first restrict expansion (14) to the photon states $|\nu\rangle = |m\rangle$ with integer number of the laser-field photons. Then we substitute $\tilde{\Psi}_\alpha^+$ into the Schrödinger equation. By acting from the left on both sides of this equation with a bra-vector $\langle n|$ and by integrating over the photon variables we obtain a chain of equations on $u_n(\tau)$:

$$(E-\hat{H}_{\rm atom}-n\omega)u_n = \langle n|\hat{H}_{\rm int}|n-1\rangle u_{n-1} + \langle n|\hat{H}_{\rm int}|n+1\rangle u_{n+1} \qquad (15)$$

If the photon occupation numbers are large $n \gg 1$ then both matrix elements on the r.h.s. of Equation (13) are equal. By using expressions (4) we arrive at a usual set of the Floquet equations:

$$(E-\hat{H}_{\rm atom}+n\omega)u_n = \frac{V}{2}(u_{n-1}+u_{n+1}) \quad , \quad n=0,\pm1\ldots, \qquad (16)$$

where $V = \boldsymbol{F}\cdot\boldsymbol{r}/2$. Thus the present formalism is equivalent to the Floquet theory within a unitary transformation.

4. Square well model

To illustrate utility of our approach we apply it to an MPI process from a one-dimensional square well. It is this model system that was considered by Burke *et al* (1991) in their seminal paper which gave rise to the spectacular success of the R-matrix Floquet theory. We consider here an electron bound initially in a square well potential $V = -2.5$ a.u. for $0 < x < 1$ and $V = 0$ for $x > 1$, with the boundary condition $R(0) = 0$ imposed on the wave functions. This potential supports only one bound state a with an energy $E = -0.4657$ a.u. We consider an MPI process when the electric field with the

frequency $\omega = 0.2$ is applied such that at least three photons are needed to ionize the system. To solve this problem we follow the steps outlined in Sec. 3. The set of equations (10) is converted into a linear system on the T-matrix elements by choosing a suitable discretization procedure for the continuous spectrum integration. We omit here details of this discretization as well as the principal value integration which are very close to that outlined in Bray (1994). In the intermediate state of Equation (10) we retained various numbers of photons. For simplicity, we count this number from the baseline of three photons in the initial state which we denote as $\alpha = |a, 3\rangle$. In the intermediate and final states we can then have negative numbers of photons. For example $n = -1$ in the final state $|\gamma\rangle$ would mean that four photons have been absorbed. Using this convention, the calculations we performed can be denoted as (-2,3), (-1,3), (0,3), (-2,4), (-1,4) and(0,4). Here (-2,3) means that the number of photons in the intermediate and final states ranges from -2 to 3.

The square well model, although extremely simple, highlights all the subtle points one encounters when applying equation (10) to a real physical system. The first problem which arises immediately is the singularity of the matrix elements $\hat{H}_{\rm int}^{\beta\alpha}$ when both states α, β lie in the continuum. In fact, one can show that for any potential for which the asymptotic behavior of the continuous wave function is given by $R_k \propto \sin(kr + \delta)$, the following result holds:

$$\int_0^\infty R_{k_i}(r) \frac{d}{dr} R_{k_f}(r)\, dr = V(k_i, k_f) + \frac{\cos(\delta_i - \delta_f)}{2} P \frac{1}{k_i - k_f}, \tag{17}$$

where $V(k_i, k_f)$ is a regular function and symbol P has a usual meaning of the principal value integral. This singularity is, of course, just a consequence of the use of the spectral expansion for the Green function and should disappear from the final result after the sum through the spectrum is carried out. A convenient way to deal with such integrals is to introduce a suitable regularization procedure. We used the following regularization formula:

$$P \frac{1}{k_i - k_f} = \lim_{\epsilon \to 0} \frac{k_i - k_f}{\epsilon^2 + (k_i - k_f)^2} \tag{18}$$

If the regularization is properly implemented the final results do not depend upon the regularization parameter ϵ. We analyze these results in the form of the partial ionization

rates computed as

$$\Gamma_n = \frac{\pi}{k_f} \left| T^{\alpha,\, k_f\, n} \right|^2 , \tag{19}$$

where the final state is specified by the momentum of a free electron k_f and the number of laser photons n. The total ionization rate is the sum of the partial rates over all the open channels.

Table 1. Convergence of the partial ionization rate Γ_3 from the (0,3) calculation with respect to the regularization parameter ϵ in Equation (18). The field strength is $F = 0.1$ a.u.

$\epsilon \times 10^2$	Γ (10^{-4} a.u.)	$\epsilon \times 10^2$	Γ (10^{-4} a.u.)
100	0.753	3.125	1.108
50	0.883	1.563	1.115
25	0.995	0.781	1.119
12.5	1.061	0.390	1.122
6.25	1.093	0.195	1.125

In Table 1 we show the partial ionization rate Γ_3 from our (0,3) calculation in which three laser photons are absorbed. The data presented in the Table clearly indicate convergence as $\epsilon \to 0$. In Table 2 we present the total ionization rates for different field strengths as computed with different numbers of laser photons (-2,3), (-1,3), (0,3), (-2,4), (-1,4) and (0,4). In these calculations the number of open channels may differ. For example, in the (-2,3) calculation one may have the final states with three, four or five photons absorbed. We remind the reader that we count the number of photons from the baseline of three photons in the initial state.

As one can see from Table 2, for small fields (up to 0.05 a.u.) all six calculations give virtually identical results. This is to be expected since for such weak fields we are in the realm of the perturbation theory. One should note, however, that even our (0,3) calculation does not correspond exactly to the third order perturbation theory. Indeed, solution of the coupled set of equations (10) amounts effectively to summation of an infinite subset of the perturbation theory terms as is illustrated in Figure 1. For small fields, as one can see from Table 2, this account of the high order terms of the perturbation theory expansion is of no importance as all six calculations give practically

Table 2. Total ionization rate Γ for different field strengths

$n^{\min}, n^{\max}$	$F = 0.025$ a.u. Γ (10^{-7} a.u.)	$F = 0.05$ a.u. Γ (10^{-5} a.u.)	$F = 0.1$ a.u. Γ (10^{-4} a.u.)
-2,3	0.1774	0.1262	1.1412
-1,3	0.1774	0.1263	1.1459
0,3	0.1751	0.1215	1.0748
-2,4	0.1766	0.1244	1.1035
-1,4	0.1766	0.1244	1.1086
0,4	0.1744	0.1198	1.0406

identical results for the ionization rate.

The situation changes somewhat for stronger fields ($F = 0.1$ a.u. in Table 2). Here the results of different calculations may vary substantially. Convergence, however, is soon achieved. The results obtained in our (-2,3) and (-2,4) calculations do not differ much. Further increase of the number of photons included in the calculation (e.g., -2,5 calculation) does not change the result appreciably. The result of the (-2,4) calculation can therefore be admitted as our final result for the total ionization rate for $F = 0.1$ a.u.

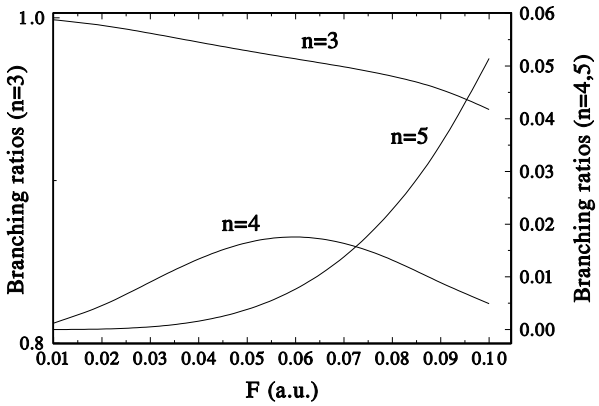

Figure 2. Branching ratios of the partial ionization rate to the total rate for various open channels in the square well model.

The partial contribution of various open channels to the total ionization rate can be judged from the data presented in Figure 2 where we plot the branching ratios for

the channels with various numbers of photons in the final state. Again, one can see that for not very large field strengths ($F < 0.05$ a.u.) the contributions of the channels with more than three photons absorbed (labeled $n = 4$ and $n = 5$ in the Figure) can be safely neglected. For larger fields their effect is becoming increasingly important.

Finally, in Figure 3, we plot the total ionization rate as a function of the field strength. We make a comparison with the calculation of Burke *et al* (1991) shown as a solid line. The dashed line in the figure indicates the third order perturbation theory. Our data deviate from the perturbation theory predictions as the field strength increases but not as much as the result of Burke *et al* (1991).

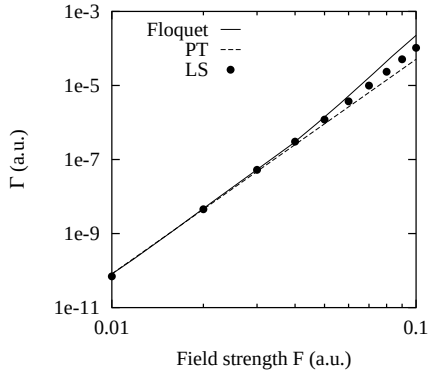

Figure 3. Total ionization rate as a function of the field strength. The solid line shows results of the R-matrix Floquet calculation of Burke *et al* (1991), the dashed line indicates the perturbation theory result. Present calculation is exhibited by points.

5. Conclusion

We developed a formalism allowing to formulate the MPI problem in terms of a set of coupled Lippmann-Schwinger equations. This is not the first attempt to describe the MPI process as a scattering phenomenon. Such a description has already been given in the literature (Bao *et al* 1996, Guo and Aberg 1988, Guo *et al* 1989, Chen *et al* 2003). We differ, however, from these works as we do not rely on the Volkov states as a set of intermediate states in our scattering theory. Instead we employ more manageable set of field free atomic states. It is of significant advantage for calculating MPI in complex atomic systems with more than one electron. This approach, however, can only be realized if one is able to generate a complete set of target states providing an accurate

quadrature rule. In this respect we can rely on the CCC method which demonstrated its ability to describe accurately singly- and doubly ionized atomic states. This gives us confidence that we can implement our method for nonperturbative description of MPI in complex atomic systems such as the helium atom.

We demonstrated utility of our approach for a model problem of a square well potential. This simple problem allowed us to test our computational scheme in which we dealt with various numerical problems, most notably, divergent matrix elements in the continuum. Much to our satisfaction, we were able to reproduce earlier results given for the square well problem by Burke *et al* (1991).

Acknowledgments

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