Keldysh-like expansion for above-threshold ionization

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(Received 17 May 1993; revised manuscript received 18 October 1993)

We develop a systematic expansion based on a semiclassical description of the ionized electrons for an atom subjected to a laser field. This expansion yields a transition amplitude for atomic multiphoton ionization which accommodates both the above-threshold ionization peaks and atomic resonances. The lowest-order term in the expansion yields the usual Keldysh approximation if the final state is replaced by a plane wave. We show that, even though this term is not reliable in general, it may produce an excellent agreement with the exact result for a three-dimensional zero-range potential and a circularly polarized field. For this situation, we calculate exactly the next nonvanishing term in the expansion, and establish precisely the conditions under which it can be neglected.

PACS number(s): 32.80.Rm, 42.50.Hz

I. INTRODUCTION

It is now well known that multiphoton processes under strong fields require, for their analysis, methods that go beyond the realm of a perturbation theory on the atom-field interaction. In fact, even though higher-order perturbation theory [1] has been successfully tested in several experiments [2], problems arise for stronger fields, due not only to the computational difficulty inherent to high-order diagrams, but also to the lack of convergence of the perturbative series. Indeed, the energy spectra of ionized electrons in recent experiments [3] display a series of peaks, corresponding to the absorption of more photons than the number strictly necessary for overcoming the binding energy. Successive peaks, even though associated with a higher number of absorptions, are comparable in magnitude, thus raising a serious concern on the applicability of perturbation theory to describe this phenomenon, commonly called *above-threshold ionization* (ATI).

Many methods, of nonperturbative nature, have been proposed to tackle this problem. Among them are adiabatic and semiclassical approximations [4], applications of Floquet theory [5], partial summation of the perturbative series [6], and dressed-state approaches [7]. Here we address ourselves to a class of methods that stem from a seminal paper by Keldysh [8–10]. These methods are based on the replacement of the final state in the time-dependent first-order transition matrix element by a Volkov state, that is, a plane wave describing the motion of the outgoing electron in the presence of an oscillating field. Depending on whether the length or the velocity gauge is used for describing the interaction between the atom and the applied laser field, this scheme is known as the Keldysh [8,9] or the Faisal-Reiss [10] ansatz, respectively. These Keldysh-Faisal-Reiss (KFR) theories have the obvious advantage of reproducing in a simple way the multiple-peaked structure of the final electronic spectrum. However, doubts have been cast on their basic foundations [11,12], and comparison of the corresponding theoretical predictions with numerical and experimental results has led to conflicting conclusions. In general, KFR models give a qualitatively good agreement with the experimentally measured relative heights of the ATI peaks, particularly so for circular polarization [13], to a lesser degree for linear polarization [14], being, however, sensitive to the choice of gauge. Keldysh-type approximations have been shown to work well for a three-dimensional short-range potential model with one bound state, in the presence of a circularly polarized field, as long as the intensity of the field is not too high [15,16]. In one dimension, on the other hand, KFR predictions are in conflict with the results of numerical simulations [11,17]. It has been argued, however, that these approximations should not be applied in one dimension at all [18]. *Total* ionization rates for long-range potentials tend to be underestimated by up to several orders of magnitude in comparison with calculations [19] based on a numerical solution of the Schrödinger equation, Floquet theory, or, where applicable, perturbation theory of lowest order. However, a correction suggested by Keldysh in his original paper [8], namely, correcting the final state for Coulomb effects, removes a good part of the discrepancy. Experimentally, total ionization rates are notoriously difficult to measure, so they do not constitute a very sensitive test of approximations, as opposed to the ATI spectrum. The only measurement [20] that can be interpreted in terms of a one-mode laser gave results that lie in between the KFR and the Floquet results and agree best with the Coulomb-corrected rate of Keldysh's original paper.

Considerable discussion has been published on the

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basic foundations of KFR theories. As we mentioned above, Keldysh's original approach was formulated in the *length gauge,* while the proposals by Faisal and Reiss were based on the *velocity gauge.* These two choices lead to different results, which has been attributed either to the fact that the approximations involved are not gauge invariant [11,12], or to the intrinsic advantages of one of the gauges over the other. Of course, there is in principle no need for an approximation to preserve the gauge invariance of the exact result. In this case, however, a natural question can be raised as to which choice leads to a better result. Similar questions arise also in other methods, as in the application of the Floquet theory by Potvliege and Shakeshaft [5], where the length gauge is shown to lead to divergences associated to the singular character at infinity of the corresponding atom-field interaction.

Most frequently, the KFR method has been viewed as a convenient ansatz, which allows a quick insight into the main dynamic effects associated with the ionization in a strong laser field, without, however, any claim to accuracy [21]. The justification of this ansatz, as well as its region of validity, have been the source of frequent debate in the literature (see, for instance, Ref. [22]).

Of course, it would be highly desirable to establish the KFR theory as the first term in a systematic approximation procedure. This would allow for a better understanding of its region of validity and, furthermore, for the calculation of successive corrections. Attempts in this direction have been presented in several papers [10,23]. In fact, it can be shown that the KFR ansatz can be obtained as the first term $M^{(0)}$ in an expansion of the exact ionization amplitude in terms of either the interatomic potential [11] or the external field interaction [12]. However, the next term $M^{(1)}$ in the expansion brings in a contribution that cancels out the KFR term. Furthermore, this contribution does not disappear by internal cancellations within $M^{(1)}$, so one cannot say that $M^{(1)}$ is smaller than $M^{(0)}$.

In this paper, we propose an alternative approach to KFR-like nonperturbative treatments. It is based on a semiclassical approximation of the final state and leads to a systematic expansion of the exact multiphoton-ionization matrix element, providing successive approximations. Our approach yields the multiple-peak ATI spectrum, and at the same time allows for transitions between atomic bound states. At the same time, it does not suffer from the above-mentioned cancellation problems of other expansions [11,12]. The first contribution of the expansion corresponds to the Keldysh amplitude (length gauge) corrected for the effects of the intra-atomic potential on the final state. Each successive approximation is expressed in terms of gauge-invariant quantities. Also, we are able to explain, using our method, the remarkable agreement between the exact result and the Keldysh expression found in the case of the three-dimensional δ-function potential [16,24].

We start our discussion in Sec. II with a brief review of the usual derivation of the KFR ansatz, in terms of an expansion of the exact transition amplitude, so that an easy comparison can be made between that method and the one we propose now. Then, in Sec. III, we introduce our approach which we apply to an explicit model in Sec. IV. In Sec. V, we summarize our conclusions.

II. KELDYSH-FAISAL-REISS ANSATZ

In this section we discuss the KFR ansatz without adopting any particular gauge, so that the discussion applies both to the length (Keldysh) and the velocity (Faisal-Reiss) gauges. The matrix element for (multi)photon ionization from an initial state $\psi_0^{(0)}(\mathbf{r}t)$ [normally the ground state of the binding potential $V(\mathbf{r})$] to a continuum state $\psi_{\mathbf{p}}^{(0)}(\mathbf{r}t)$, still in the presence of the binding potential, with asymptotic momentum $\mathbf{p}$, is given by

$$M_{\mathbf{p}}=\lim_{\substack{t\to\infty\\ t'\to-\infty}}\int d^3r\,d^3r'\psi_{\mathbf{p}}^{(0)}(\mathbf{r}t)^*iG(\mathbf{r}t,\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\ . \tag{2.1}$$

Here $G(\mathbf{r}t,\mathbf{r}'t')$ is the retarded propagator in the presence of both the external time-dependent field which causes the ionization and the atomic binding potential $V(\mathbf{r})$. It satisfies

$$G(\mathbf{r}t,\mathbf{r}'t')=G_0(\mathbf{r}t,\mathbf{r}'t')+\int d^3r''dt''[G_0(\mathbf{r}t,\mathbf{r}''t'')H_I(\mathbf{r}''t'')G(\mathbf{r}''t'',\mathbf{r}'t')]\ , \tag{2.2}$$

or, symbolically,

$$G=G_0+G_0H_IG\ . \tag{2.2'}$$

In this expression G_0 denotes the retarded propagator in the presence of the binding potential only:

$$iG_0(\mathbf{r}t,\mathbf{r}'t')=\Theta(t-t')\left[\sum_n\psi_n^{(0)}(\mathbf{r}t)\psi_n^{(0)}(\mathbf{r}'t')^*+\int d^3p\ \psi_{\mathbf{p}}^{(0)}(\mathbf{r}t)\psi_p^{(0)}(\mathbf{r}'t')^*\right]\ , \tag{2.3}$$

where the sum and the integral are over the bound states and continuum states of the potential $V(\mathbf{r})$, respectively. The wave functions in Eq. (2.1) [as well as (2.3)] are those in the absence of the external field. We assume that this field is turned off for early and late times ($t'\to-\infty$, $t\to\infty$). The matrix element (2.1) is then evidently gauge invariant. The interaction H_I represents the interaction between the atom and the external field. We assume that the long-wavelength approximation is justified for the external field so that its vector potential $\mathbf{A}(t)$ depends only on time.

Another equation satisfied by $G(\mathbf{r}t,\mathbf{r}'t')$ is

$$G=G'+G'VG\ , \tag{2.4}$$

where now G' is the retarded propagator in the absence of the binding potential, with only the external field present.

If we insert (2.4) into (2.1), we get

$$M_{\mathrm{p}}=\lim_{\substack{t\to\infty\\ t'\to-\infty}}\left[\int d^3r\,d^3r'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*iG'(\mathbf{r}t,\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\right.$$
$$\left.+i\int d^3r\,d^3r'd^3r''dt''\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*G'(\mathbf{r}t,\mathbf{r}''t'')V(\mathbf{r}'')G(\mathbf{r}''t'',\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\right]\ . \tag{2.5}$$

Here the first term on the right-hand side (rhs) is zero in the limit where $t\to\infty$ and $t'\to-\infty$. In this limit the propagator G' reduces to the free propagator (*not* to the propagator G_0 in the presence of the binding potential) and, loosely speaking, free propagation cannot lift the initially bound particle up into the continuum. (The argument can be made rigorous by just inserting a plane-wave expansion of the free propagator.) In the remaining term in Eq. (2.5) we replace G by G_0, viz., the first term on the rhs of Eq. (2.2). We then get

$$M_{\mathrm{p}}\to M_{\mathrm{p},0}=\lim_{t\to\infty}\int d^3r\,d^3r'dt'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*G'(\mathbf{r}t,\mathbf{r}'t')$$
$$\times V(\mathbf{r}')\psi_0^{(0)}(\mathbf{r}'t')\ . \tag{2.6}$$

Upon replacing the final state by a momentum eigenfunction, one gets precisely the expression proposed in Ref. [9] (where the length gauge was used):

$$M'_{\mathrm{p},0}=-i\int d^3r\,dt\,\psi_{\mathrm{p}}^{(V)}(\mathbf{r}t)^*V(\mathbf{r})\psi_0^{(0)}(\mathbf{r}t)\ . \tag{2.7}$$

On the other hand, with the help of Eq. (2.4), in the form $G'V=1-G'G^{-1}$, and using $G^{-1}=G_0^{-1}-H_I$, we can also rewrite $M_{\mathrm{p}}^{(0)}$ as [11]

$$M_{\mathrm{p},0}=\lim_{t\to\infty}\int d^3r\,d^3r'dt'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*G'(\mathbf{r}t,\mathbf{r}'t')$$
$$\times H_I(\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\ . \tag{2.8}$$

Again replacing the final state by a momentum eigenfunction, one gets precisely the expression usually associated with the KFR ansatz [8,10]:

$$M'_{\mathrm{p},0}=-i\int d^3r\,dt\,\psi_{\mathrm{p}}^{(V)}(\mathbf{r}t)^*H_I(\mathbf{r}t)\psi_0^{(0)}(\mathbf{r}t)\ , \tag{2.9}$$

with

$$\psi_{\mathrm{p}}^{(V)}(\mathbf{r}'t')^*=\lim_{t\to\infty}\int d^3r(2\pi)^{-3/2}e^{-i\mathbf{p}\cdot\mathbf{r}}e^{i(\mathbf{p}^2/2m)t}$$
$$\times iG'(\mathbf{r}t,\mathbf{r}'t') \tag{2.10}$$

the Volkov solution. The process is described as a transition, induced by the interaction H_I, between the initial bound state and a Volkov state, that is, a plane-wave solution of the Schrödinger equation for an electron interacting with a plane electromagnetic wave.

The above discussion helps one to understand some of the basic ingredients of the KFR ansatz. The approximation of the final state by a plane wave might be valid for sufficiently short-range potentials and highly energetic electrons. It should certainly not be used for the calculation of the total ionization rate, since then an integration over all final momenta should be performed, and low momenta contribute appreciably to the result [11]. One should note that this approximation is not necessary and has actually been circumvented in the literature by keeping the final state as an eigenstate of the atomic Hamiltonian, conveniently modified by the external laser field [11,25,26]. On the other hand, approximating G by G_0 in (2.5) amounts to neglecting the effects of the field on the initial bound state. In particular, the depletion of this state due to ionization and transitions to other bound states, as well as the level shift due to the applied field, are disregarded. Neglecting these effects would be, at first sight, a weak-field approximation, which in particular would not account for atomic resonances. In order to assess the validity of this approximation, however, one would have to show that, for weak fields, the remaining terms in the expansion are smaller in magnitude than the Keldysh term.

The procedure described above actually allows one to calculate the corrections to the KFR ansatz, which come from further iterations of Eq. (2.2). We avoid here the approximation of the final state by a plane wave, and concentrate on the approximation which results from the truncation of the iterative series stemming from (2.2). The first correction is

$$M_{\mathrm{p},1}=\lim_{\substack{t\to\infty\\ t'=-\infty}}i\int d^3r\,d^3r'd^3r''dt''d^3r'''dt'''\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*$$
$$\times G'(\mathbf{r}t,\mathbf{r}''t'')V(\mathbf{r}'')G_0(\mathbf{r}''t'',\mathbf{r}'''t''')H_I(\mathbf{r}'''t''')G_0(\mathbf{r}'''t''',\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\ . \tag{2.11}$$

The propagator G' satisfies (symbolically)

$$(G_0^{-1}+V-H_I)G'=G'(G_0^{-1}+V-H_I)=1 \tag{2.12}$$

so that

$$G'VG_0=G'H_IG_0+G_0-G'\ . \tag{2.13}$$

Using Eq. (2.13) in Eq. (2.11) we obtain

$$M_{\mathrm{p},1}=-\lim_{t\to\infty}\int d^3r\,d^3r'dt'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*G'(\mathbf{r}t,\mathbf{r}'t')H_I(\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')-i\int d^3r\,dt\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*H_I(\mathbf{r}t)\psi_0^{(0)}(\mathbf{r}t)$$
$$+\lim_{t\to\infty}\int d^3r\,d^3r'd^3r''dt'dt''\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*G'(\mathbf{r}t,\mathbf{r}'t')H_I(\mathbf{r}'t')G_0(\mathbf{r}'t',\mathbf{r}''t'')H_I(\mathbf{r}''t'')\psi_0^{(0)}(\mathbf{r}''t'')\ . \tag{2.14}$$

Comparison of this expression with (2.9) shows that the KFR ansatz is exactly canceled out by the first contribution on the right-hand side of (2.14). It is also clear that in general the contributions from the first and the third terms on the right-hand side of this expression are quite different, since the third term involves one more transition between atomic states than the first one (in particular, the third term gives rise to resonances which are not present in the first term). However, one could still hope that the difference between the first two terms on the right-hand side of (2.14) is of higher order in H_I, so that (2.7) would be a bona fide lowest-order term of a consistent expansion. This would only be true in general if G' approached G_0 when $H_I\to 0$, which is certainly not true: in this limit G' approaches the free-particle propagator, while G_0 remains the free-atom propagator. Therefore the correction $M_{\mathrm{p},1}$ is not necessarily smaller than the KFR term.

Examining more closely the structure of expression (2.14), we see that the second term on the right-hand side is just the ordinary perturbation-theory contribution, associated with one-photon transitions, while the last term is a higher-order KFR-like contribution, equally canceled when the next correction is considered. For a multiphoton transition (multiplicity greater than one), the second term will actually not contribute at all due to energy conservation. As the expansion is continued, however, one would eventually pick up the perturbation-theory contribution, which would become relevant in the term of the same order as the multiplicity of the transition.

We show now that it is actually possible to replace the KFR ansatz by a closely related one, which can be shown to be the lowest-order contribution of a consistent expansion, such that the next term indeed becomes of higher order when the external-field amplitude goes to zero.

III. SEMICLASSICAL ASYMPTOTICS: A NEW EXPANSION METHOD

Our proposal is based on the consideration that, after leaving the atom, the electron undergoes a quasiclassical motion. Henceforth we will in all gauge-dependent quantities specify the gauge by a superscript. We find it convenient to start from the $\mathbf{p}\cdot\mathbf{A}$ gauge so that

$$H_I(\mathbf{r}t)\equiv H_I^{(A)}(\mathbf{r}t)=-\frac{e}{m}\hat{\mathbf{p}}'\cdot\mathbf{A}(t)+\frac{e^2}{2m}\mathbf{A}^2(t)\ , \tag{3.1}$$

with $\hat{\mathbf{p}}=-i\nabla$.

We now decompose the interaction Hamiltonian (3.1) into a c-number part and the remainder according to

$$H_I^{(A)}(\mathbf{r}t)=H_{I,\mathbf{p}}^{(A,\mathrm{cl})}(t)+\tilde{H}_{I,\mathbf{p}}^{(A)}(\mathbf{r}t)\ , \tag{3.2}$$

with

$$H_{I,\mathbf{p}}^{(A,\mathrm{cl})}(t)=-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(t)+\frac{e^2}{2m}\mathbf{A}^2(t)\ , \tag{3.3}$$

$$\tilde{H}_{I,\mathbf{p}}^{(A)}(\mathbf{r}t)=-\frac{e}{m}(\hat{\mathbf{p}}-\mathbf{p})\cdot\mathbf{A}(t)\ . \tag{3.4}$$

The c-number momentum $\mathbf{p}$ (notice the distinction between $\mathbf{p}$ and $\hat{\mathbf{p}}$) is at this point arbitrary. Later, however, we will identify it with the asymptotic momentum of the continuum state under consideration. A similar decomposition was used in the treatment [27] of infrared divergences of relativistic quantum electrodynamics. Now, if we insert the decomposition (3.2) into Eq. (2.2) for the complete propagator chosen now in the $\mathbf{p}\cdot\mathbf{A}$ gauge it turns out that the c-number part (3.3) can be entangled with G_0 so that, in the symbolic notation introduced in Eq. (2.2′),

$$G^{(A)}=\tilde{G}^{(A)}+\tilde{G}^{(A)}\tilde{H}_{I,\mathbf{p}}^{(A)}G^{(A)}\ , \tag{3.5}$$

with

$$\tilde{G}^{(A)}(\mathbf{r}t,\mathbf{r}'t')=\exp\left[-i\int_{t'}^{t}d\tau\,H_{I,\mathbf{p}}^{(A,\mathrm{cl})}(\tau)\right]G_0(\mathbf{r}t,\mathbf{r}'t')\ . \tag{3.6}$$

That is, if the propagator G_0 in the presence of the binding potential is known, then so is $\tilde{G}^{(A)}$.

Let us now, in the integral expression (2.1) for the matrix element M_{p}, iterate the full propagator once using Eq. (3.5). The first term cancels in view of the orthogonality of $\psi_0^{(0)}$ and $\psi_{\mathrm{p}}^{(0)}$, which is not obstructed by the newly introduced phase in Eq. (3.6). We obtain

$$M_{\mathrm{p}}=\lim_{t'\to-\infty}\int d^3r\,dt\,d^3r'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*\Phi_{\mathrm{p}}(t)\tilde{H}_{I,\mathbf{p}}^{(A)}(\mathbf{r}t)$$
$$\times G^{(A)}(\mathbf{r}t,\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t')\ , \tag{3.7}$$

where

$$\Phi_{\mathrm{p}}(t)=\exp\left[-i\int_{t}^{\infty}d\tau\,H_{I,\mathbf{p}}^{(A,\mathrm{cl})}(\tau)\right]\ . \tag{3.8}$$

Equation (3.7) is still exact. Notice that the matrix element M_{p} becomes zero if $\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)$ is replaced by a plane wave with momentum $\mathbf{p}$ since

$$\tilde{H}_{I,\mathbf{p}}^{(A)}(\mathbf{r}t)e^{i\mathbf{p}\cdot\mathbf{r}}=0\ . \tag{3.9}$$

There is no contradiction here. Recall that in the derivation of Eq. (3.7) we employed the fact that $\psi_{\mathrm{p}}^{(0)}$ and $\psi_0^{(0)}$ are states in the presence of the same binding potential $V(\mathbf{r})$. Hence replacing $\psi_{\mathrm{p}}^{(0)}$ by a plane wave in (3.7) is not equivalent to making the same replacement in (2.1).

Getting manageable approximations out of Eq. (3.7) depends on approximating the propagator $G^{(A)}$. Before doing so we transform to the electric-field gauge,

$$G^{(A)}(\mathbf{r}t,\mathbf{r}'t')=e^{ie\mathbf{r}\cdot\mathbf{A}(t)}G^{(E)}(\mathbf{r}t,\mathbf{r}'t')e^{-ie\mathbf{r}'\cdot\mathbf{A}(t')} , \tag{3.10}$$

where $G^{(E)}$ satisfies Eq. (2.2):

$$G^{(E)}=G_0+G_0H_I^{(E)}G^{(E)}=G_0+G^{(E)}H_I^{(E)}G_0 , \tag{3.11}$$

with

$$H_I(\mathbf{r},t)\equiv H_I^{(E)}(\mathbf{r}t)=-e\mathbf{r}\cdot\mathbf{E}(t) . \tag{3.12}$$

We now have

$$M_{\mathrm{p}}=\lim_{t'\to\infty}\int d^3r\,dt\,d^3r'\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*\Phi_{\mathrm{p}}(t) \times\tilde{H}_{I,\mathrm{p}}^{(A)}(\mathbf{r}t)\exp[ie\mathbf{r}\cdot\mathbf{A}(t)] \times G^{(E)}(\mathbf{r}t,\mathbf{r}'t')\psi_0^{(0)}(\mathbf{r}'t') . \tag{3.13}$$

At this point it is reasonable to build an approximation scheme on iterating $G^{(E)}$ with the help of Eq. (3.11). The interaction of the electron in the continuum with the external field is fairly well described by the phase $\Phi_{\mathrm{p}}(t)$, which is the Volkov phase, while inside the atom a perturbation expansion in terms of $\mathbf{r}\cdot\mathbf{E}$ provides optimal convergence. Notice that this is not so for a perturbation expansion based on $H_I^{(A)}$ [Eq. (3.1)]. First, in the latter case many more states must be considered in sums over inserted intermediate states, in order to achieve satisfactory convergence, since the proportionality of the matrix element of $\mathbf{p}\cdot\mathbf{A}$ to the energy difference between the connected states enhances the contribution of farther-lying states; and, second, there are extensive cancellations between the $\mathbf{p}\cdot\mathbf{A}$ and the $\mathbf{A}^2$ terms, which would make any evaluation beyond the lowest-order extremely cumbersome.

Carrying out the suggested iteration we obtain

$$M_{\mathrm{p}}=\sum_{n=0}^{\infty}M_{\mathrm{p}}^{(n)} , \tag{3.14}$$

where

$$M_{\mathrm{p}}^{(n)}=-i\psi_{\mathrm{p}}^{(0)*}\Phi_{\mathrm{p}}\tilde{H}_{I,\mathrm{p}}^{(A)}\exp(ie\mathbf{r}\cdot\mathbf{A})(G_0H_I^{(E)})^n\psi_0^{(0)} , \tag{3.15}$$

again in symbolic notation. The lowest-order term of this expansion, viz.,

$$M_{\mathrm{p}}^{(0)}=-i\int d^3r\,dt\,\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*\Phi_{\mathrm{p}}(t) \times\left[-\frac{e}{m}(\hat{\mathbf{p}}-\mathbf{p})\cdot\mathbf{A}(t)\right] \times\exp[ie\mathbf{r}\cdot\mathbf{A}(t)]\psi_0^{(0)}(\mathbf{r}t) , \tag{3.16}$$

can be rewritten as follows. Notice that

$$-\frac{e}{m}(\hat{\mathbf{p}}-\mathbf{p})\cdot\mathbf{A}(t)\Phi_{\mathrm{p}}(t)=\left[i\frac{\partial}{\partial t}+\frac{1}{2m}[\hat{\mathbf{p}}-e\,\mathbf{A}(t)]^2-\frac{1}{2m}\hat{\mathbf{p}}^2\right]\Phi_{\mathrm{p}}(t) , \tag{3.17}$$

and

$$i\frac{\partial}{\partial t}[e^{ie\mathbf{r}\cdot\mathbf{A}(t)}\psi_0^{(0)}(\mathbf{r}t)]=\left[\frac{(\hat{\mathbf{p}}-e\,\mathbf{A})^2}{2m}+V(\mathbf{r})+e\mathbf{r}\cdot\mathbf{E}(t)\right] \times e^{ie\mathbf{r}\cdot\mathbf{A}(t)}\psi_0^{(0)}(\mathbf{r}t) . \tag{3.18}$$

Using Eq. (3.17) in Eq. (3.16), integrating by parts, and using Eq. (3.18), we arrive at

$$M_{\mathrm{p}}^{(0)}=-i\int d^3r\,dt\,\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)^*\Phi_{\mathrm{p}}(t)e^{ie\mathbf{r}\cdot\mathbf{A}(t)} \times[-e\mathbf{r}\cdot\mathbf{E}(t)]\psi_0^{(0)}(\mathbf{r}t) . \tag{3.19}$$

The integration by parts does not yield any boundary terms as we assumed that $\mathbf{A}(t)\to\mathbf{0}$ for $t\to\pm\infty$.

Equations (3.16) and (3.19) provide equivalent expressions for the matrix element $M_{\mathrm{p}}^{(0)}$. However, the equivalence only holds if both $\psi_{\mathrm{p}}^{(0)}$ and $\psi_0^{(0)}$ are wave functions corresponding to the same potential $V(\mathbf{r})$, a fact that was used in deriving Eq. (3.19) from Eq. (3.16). If $\psi_{\mathrm{p}}^{(0)}$ is replaced by a plane wave, this equivalence is destroyed: Eq. (3.16) then yields zero while Eq. (3.19) in general will not. If $\psi_{\mathrm{p}}^{(0)}$ is expanded in a Born series, then the first (plane-wave) term will not contribute to Eq. (3.16), and the leading contribution will come from the once-iterated term containing the potential. On the other hand, the plane-wave term may make the dominant contribution to Eq. (3.19). In fact, replacing $\psi_{\mathrm{p}}^{(0)}$ by a plane wave in Eq. (3.19) will actually be exact if the initial and the scattered part of the final state are s waves and the field is circularly polarized. This can be easily shown by considering that, for a circularly polarized field, $\mathbf{A}\cdot\mathbf{E}=\mathbf{0}$. Therefore, changing to a reference frame which rotates with the field vector and choosing the y axis parallel to the electric field, we can see that the integrand in Eq. (3.19) is odd with respect to the transformation $y\to-y$, if we replace $\psi_{\mathrm{p}}^{(0)}(\mathbf{r}t)$ by the scattered part of the final state, and this is assumed to be an s wave, in the same way as the initial state. Therefore under these conditions only the plane-wave part of the final state contributes to Eq. (3.19). This will be the case for the three-dimensional δ-function potential, discussed in the next section: this potential has only one $l=0$ bound state, and, due to its zero range, scatters only s waves.

The consequences of replacing $\psi_{\mathrm{p}}^{(0)}$ by a plane wave have been investigated in detail for the case of one-photon ionization in ordinary perturbation theory [28]. It has been shown that owing to this replacement the originally gauge-independent matrix element developed a spurious gauge dependence. For a Coulomb potential, the result in the length gauge is larger than in the velocity gauge by a factor of 2, with the latter approaching the exact result in the high-energy limit. The discrepancy remains and can even get worse for short-range potentials. Thus, if the potential is regular at $r=0$, the length matrix element is three times larger than the velocity matrix element at high energies [28]. For a zero-range potential, however, both gauges yield identical results [this

can easily be shown from Eq. (2) of Ref. [28] or from our Eq. (2.7)]. Within the Floquet approach and in the weak-field limit, Potvliege and Shakeshaft [5] have demonstrated that the plane-wave approximation leads to discrepancies which stay in most cases, but not always, within one order of magnitude.

The amplitude $M_p^{(0)}$ as given in Eq. (3.19) (as well as the higher approximants $M_p^{(n)}$ to be derived below) is manifestly gauge independent since, through $\Phi_p(t)$ and the product $\psi_p^{(0)}(\mathbf{r},t)^*\exp(ie\mathbf{r}\cdot\mathbf{A})$, it only depends on the gauge-invariant mechanical momentum $\pi(t)=\mathbf{p}-e\,\mathbf{A}(t)$, as well as the gauge-invariant quantities $\mathbf{r}$ and $\mathbf{E}$. However, upon replacing $\psi_p^{(0)}$ by a plane wave, it will, for a Coulomb potential and one-photon ionization in the low-intensity and high-energy limit, deviate by a factor of 2 from the exact result, as discussed in the preceding paragraph.

It should be remarked that Eq. (3.19) with the exact wave functions has been used, in an *ad hoc* fashion, for extensive calculations of above-threshold ionization in hydrogen [25,26].

We now want to rewrite

$$M_p^{(1)}=-i\int d^3r\,dt\,d^3r'dt'\psi_p^{(0)}(\mathbf{r}t)^*\Phi_p(t)\left[-\frac{e}{m}(\hat{\mathbf{p}}-\mathbf{p})\cdot\mathbf{A}(t)\right]\exp[ie\mathbf{r}\cdot\mathbf{A}(t)]G_0(\mathbf{r}t,\mathbf{r}'t')[-e\mathbf{r}'\cdot\mathbf{E}(t')]\psi_0^{(0)}(\mathbf{r}'t') \quad (3.20)$$

along the same lines. We start as before with Eq. (3.15), and use

$$i\frac{\partial}{\partial t}e^{ie\mathbf{r}\cdot\mathbf{A}(t)}G_0(\mathbf{r}t,\mathbf{r}'t')=\delta(t-t')\delta(\mathbf{r}-\mathbf{r}')e^{ie\mathbf{r}\cdot\mathbf{A}(t)}+\left[\frac{1}{2m}(\hat{\mathbf{p}}-e\,\mathbf{A})^2+V(\mathbf{r})+e\mathbf{r}\cdot\mathbf{E}(t)\right]e^{ie\mathbf{r}\cdot\mathbf{A}(t)}G_0(\mathbf{r}t,\mathbf{r}'t') \quad (3.21)$$

in place of Eq. (3.18). This yields

$$\begin{aligned}M_p^{(1)}=&-i\int d^3r\,dt\,d^3r'dt'\psi_p^{(0)}(\mathbf{r}t)^*\Phi_p(t)\exp[ie\mathbf{r}\cdot\mathbf{A}(t)][-e\mathbf{r}\cdot\mathbf{E}(t)]G_0(\mathbf{r}t,\mathbf{r}'t')[-e\mathbf{r}'\cdot\mathbf{E}(t')]\psi_0^{(0)}(\mathbf{r}'t')\\&+i\int d^3r\,dt\,\psi_p^{(0)}(\mathbf{r}t)^*\Phi_p(t)\exp[ie\mathbf{r}\cdot\mathbf{A}(t)][-e\mathbf{r}\cdot\mathbf{E}(t)]\psi_0^{(0)}(\mathbf{r}t)\\&+\int d^3r\,d^3r'dt'\left[\lim_{t\to\infty}-\lim_{t\to-\infty}\right]\psi_p^{(0)}(\mathbf{r}t)^*\Phi_p(t)\exp[ie\mathbf{r}\cdot\mathbf{A}(t)]G_0(\mathbf{r}t,\mathbf{r}'t')[-e\mathbf{r}'\cdot\mathbf{E}(t')]\psi_0^{(0)}(\mathbf{r}'t')\,. \quad (3.22)\end{aligned}$$

Here the boundary term at $t\to\infty$ makes a contribution while the one at $t\to-\infty$ does not since the propagator G_0 is retarded and $\mathbf{E}(t)\to0$ for $t\to-\infty$. All of the $M_p^{(n)}$ with $n\geq1$ can be rewritten in exactly the same way. We may summarize the result as follows:

$$M_p^{(0)}=K_p^{(0)}\,, \quad (3.23)$$

$$M_p^{(n)}=K_p^{(n)}-K_p^{(n-1)}+P_p^{(n)}\quad(n\geq1)\,, \quad (3.24)$$

where, in symbolic notation,

$$\begin{aligned}K_p^{(n)}=&-i\psi_p^{(0)*}\Phi_p e^{ie\mathbf{r}\cdot\mathbf{A}}(-e\mathbf{r}\cdot\mathbf{E})\\&\times[G_0(-e\mathbf{r}\cdot\mathbf{E})]^n\psi_0^{(0)}\end{aligned} \quad (3.25)$$

and

$$P_p^{(n)}=-i\psi_p^{(0)*}(-e\mathbf{r}\cdot\mathbf{E})[G_0(-e\mathbf{r}\cdot\mathbf{E})]^{n-1}\psi_0^{(0)}\,. \quad (3.26)$$

The $P_p^{(n)}$ are just the matrix elements of ordinary nth order perturbation theory in $\mathbf{r}\cdot\mathbf{E}$. If a minimum of N photons are required for ionization then

$$P_p^{(n)}\equiv0\quad\text{for } n<N\,. \quad (3.27)$$

$K_p^{(0)}$ coincides precisely with the Keldysh ansatz [8] (but *not with the Faisal-Reiss expression*), if $\psi_p^{(0)}$ is replaced by a plane wave. Of course, this approximation has the problems mentioned in Sec. II and below Eq. (3.19). Note that the phase $\exp(ie\,\mathbf{A}\cdot\mathbf{r})$ combines with the factor $\exp(-i\mathbf{p}\cdot\mathbf{r})$ coming from the final plane-wave state to yield a dependence of the matrix element on the kinetic momentum, a gauge-invariant quantity. Therefore this phase factor should not be neglected, and it has indeed been shown to alter results significantly for higher intensities [26]. Potvliege and Shakeshaft [5] arrived at the same conclusion, even in the weak-field limit, in the context of the Floquet method. The presence of the phase $\exp(ie\,\mathbf{A}\cdot\mathbf{r})$ also helps in explaining the experimental absence of a fourfold symmetry in the ATI angular distribution for general elliptic polarization (which is predicted by $K_p^{(0)}$ without the phase factor) [29].

Equations (3.23) and (3.24) have a remarkable property, similar to the one discussed in connection with the expansion in Sec. II. It is clear from (3.24) that each successive term in the iterative expansion of M_p [Eq. (3.13)] brings in two Keldysh-like contributions, one which cancels out the result of the previous iteration, and the other which represents a higher-order Keldysh term. Again, it is clear that the contribution from $K_p^{(n)}$ will be in general quite different from the one coming from $K_p^{(n-1)}$, since they involve a different number of atomic propagators [in particular, $K_p^{(n)}$ exhibits more singular denominators, associated with resonances, than $K_p^{(n-1)}$]. Contrary, however, to what happens in the expansion discussed in Sec. II, we can see from (3.24)–(3.26) that this time $K_p^{(n-1)}$ does cancel out $P_p^{(n)}$ when the external field goes to zero. This has the following interesting implication: If we take (3.27) into consideration, we have, for $m<N$,

$$\sum_{n=0}^{m}M_p^{(n)}=K_p^{(m)}\quad(m<N)\,, \quad (3.28)$$

while for $m\geq N$ we have

$$\sum_{n=0}^{m} M_p^{(n)} = K_p^{(m)} + \sum_{n=N}^{m} P_p^{(n)} \ . \tag{3.29}$$

From these two equations, and from the above discussion, we can see that, while for $m < N$ it is not generally true that $M_p^{(m+1)}$ is smaller than $M_p^{(m)}$, for $m \geq N$, on the other hand, one can be sure that for sufficiently weak fields $M_p^{(m)}$ is indeed of higher order than $M_p^{(m-1)}$. This reasoning indicates that $M_p^{(N-1)}$ is the most reliable Keldysh-like approximation. It has the appealing feature of describing the Nth order transition between atomic states, and at the same time it takes into account the semiclassical change in the final state due to the applied field, which provides the complete ATI spectrum. We note that this contribution has the same general structure as the ansatz proposed in Ref. [30], which has provided good agreement with experimental results (cf. Ref. [14]).

It is clear also that if one goes one term beyond this approximation, ordinary perturbation theory starts to play a role. That is, for $m \geq N$ (N being the multiphoton order of the ionization process), term by term ordinary $\mathbf{r}\cdot\mathbf{E}$ perturbation theory creeps back in and replaces everything that has been done before. Ultimately

$$\lim_{m\to\infty} \sum_{n=0}^{m} M_p^{(n)} = \lim_{m\to\infty} \sum_{n=N}^{m} P_p^{(n)} \ , \tag{3.30}$$

as one should expect, provided both sums converge. Of course, for some special cases, the first term in the expansion, $K_p^{(0)}$, may already yield an excellent approximation. Physically, this would be the situation whenever multiphoton transitions do not involve bound states other than the ground state. One such case is the three-dimensional δ-function potential to be discussed below and possibly any other short-range potential with just one bound state. It is quite possible that more general potentials may be well described, at least qualitatively, by $K_p^{(0)}$, provided that intermediate resonances play no role. The latter question is strongly dependent on the polarization of the external field, since for circular polarization resonances are less likely to be encountered than for linear polarization. If an n-photon resonance is close, one could expect that $K_p^{(n)}$ should be used instead. In fact, one can do better than that. When intermediate resonances become important, one can sum up, in the expansion (3.14) and (3.15), the terms which contain small denominators. That is, we apply to (3.14) the same summation techniques used in conventional perturbation theory to deal with resonances [6]. This would yield a corrected expression for $M_p^{(n)}$ in (3.15) in which the small denominators are replaced by expressions containing linewidths. We can now apply to these corrected contributions the procedure leading to Eqs. (3.23)–(3.26), in which all (intermediate) G_0's with small denominators are replaced by resonant expressions [note that the leftmost G_0 in Eq. (3.15) is not affected by this summation, and therefore the partial integrations leading from (3.15) to (3.24) can still be performed; cf. Eq. (3.21)]. This approach yields an expression for $K_p^{(N)}$ that accommodates both the ATI peaks and the intermediate resonances. It is interesting to note that our procedure actually leads to the justification of a modified "resonant ansatz" introduced already by Keldysh in Ref. [8].

IV. EXAMPLE

In this section we will compute the lowest approximant $M_p^{(0)}$ for the three-dimensional δ-function model [16,24], discuss the validity of this approximation in this case, and compare it with the standard Keldysh approximation $M_p^{(K)}$.

The model is defined by the zero-range potential

$$V(\mathbf{r}) = \frac{2\pi}{m\kappa}\delta(\mathbf{r})\frac{\partial}{\partial r}r \ , \tag{4.1}$$

which has a single bound state at energy

$$E_0 = -\frac{\kappa^2}{2m} \ . \tag{4.2}$$

A complete orthonormal set of wave functions consists of

$$\psi_0^{(0)}(\mathbf{r}t) = \frac{\sqrt{\kappa}}{2\pi}\frac{e^{-\kappa r}}{r}e^{i|E_0|t} \tag{4.3}$$

for the bound state and

$$\psi_p^{(0)}(\mathbf{r}t) = \frac{1}{(2\pi)^{3/2}}\left[e^{i\mathbf{p}\cdot\mathbf{r}} - \frac{1}{\kappa+ip}\frac{e^{ipr}}{r}\right]e^{-iE_p t} \tag{4.4}$$

for the continuum states, where $E_p = \mathbf{p}^2/2m$. We will now evaluate the matrix elements $M_p^{(n)}$ for this potential and an external field specified by the vector potential $\mathbf{A}(t)$.

We start with $M_p^{(0)}$ in the form (3.16). Inserting the wave functions (4.3) and (4.4) and replacing $(e/m)\mathbf{p}\cdot\mathbf{A}$ in the integral by a time derivative acting on $\Phi_p(t)$ yields

$$M_p^{(0)} = \frac{i\sqrt{\kappa}}{(2\pi)^2(\kappa-ip)}\int d^3r\,dt\frac{e^{-ipr}}{r}e^{i(E_p+|E_0|)t}\exp\left[-i\frac{e^2}{2m}\int_t^\infty d\tau\,\mathbf{A}^2(\tau)\right]$$
$$\times\left[i\frac{e}{m}\mathbf{A}\cdot\nabla + i\frac{\partial}{\partial t}\right]e^{ie\mathbf{A}\cdot\mathbf{r}}\frac{e^{-\kappa r}}{r}\exp\left[i\frac{e}{m}\int_t^\infty d\tau\,\mathbf{p}\cdot\mathbf{A}(\tau)\right] \ . \tag{4.5}$$

Integrating by parts with respect to t and having one-half of the derivative $\partial/\partial x$ act to the right while integrating by parts with respect to the remaining half, the term in square brackets is replaced by

$$E_p + |E_0| - \frac{ie}{2m}(\kappa-ip)\frac{\mathbf{A}\cdot\mathbf{r}}{r} - e\mathbf{r}\cdot\mathbf{E} \ , \tag{4.6}$$

where $\mathbf{E} = -\partial\mathbf{A}/\partial t$. We notice that

$$\frac{e\,\mathbf{A}\cdot\mathbf{r}}{r}e^{ie\,\mathbf{A}\cdot\mathbf{r}}=-i\frac{\partial}{\partial r}e^{ier[(\mathbf{A}\cdot\mathbf{r})/r]}\ . \tag{4.7}$$

Another integration by parts with respect to r leads to

$$M_{\mathrm{p}}^{(0)}=i\frac{\sqrt{\kappa}}{(2\pi)^2(\kappa-ip)}\int dt\left\{\int\frac{d^3r}{r^2}e^{-(\kappa+ip)r}\exp i[(E_{\mathrm{p}}+|E_0|)t]\Phi_{\mathrm{p}}(t)[-e\mathbf{r}\cdot\mathbf{E}(t)]e^{ier\cdot\mathbf{A}(t)}\right.$$
$$\left.+\frac{2\pi}{m}(\kappa-ip)\exp[i(\mathbf{E}_{\mathrm{p}}+|E_0|)t]\Phi_{\mathrm{p}}(t)\right\}\ . \tag{4.8}$$

The second term on the rhs of Eq. (4.8) is a boundary term at $r=0$ originating from the integration by parts. The spatial integration in the first term yields

$$\int\frac{d^3r}{r^2}e^{-(\kappa+ip)r}(-e\mathbf{r}\cdot\mathbf{E})e^{ier\cdot\mathbf{A}}=\frac{2\pi}{e}\frac{\mathbf{A}\cdot\mathbf{E}}{\mathbf{A}^2}\left[-\frac{1}{|\mathbf{A}|}\ln\frac{\kappa+ip+ie|\mathbf{A}|}{\kappa+ip-ie|\mathbf{A}|}+2ie\frac{\kappa+ip}{(\kappa+ip)^2+e^2\mathbf{A}^2}\right]$$
$$=-\frac{8\pi i}{3}\frac{e^2\mathbf{A}\cdot\mathbf{E}}{(\kappa+ip)^3}\left\{1+0\left[\frac{e^2\mathbf{A}^2}{(\kappa+ip)^2}\right]\right\}\ . \tag{4.9}$$

Hence the first term does not contribute for circular polarization where $\mathbf{A}\cdot\mathbf{E}=0$.

The second term in Eq. (4.8), viz., the boundary term

$$M_{\mathrm{p,BT}}^{(0)}=i\frac{\sqrt{\kappa}}{2\pi m}\int_{-\infty}^{\infty}dt\,\exp[i(E_{\mathrm{p}}+|E_0|)t]\Phi_{\mathrm{p}}(t)$$
$$=\frac{i\sqrt{\kappa}}{2\pi m}\int_{-\infty}^{\infty}dt\,\exp(i|E_0|t)$$
$$\times\exp\left[\frac{i}{2m}\int^t dt\,\pi(t)^2\right]\ , \tag{4.10}$$

is easily seen to be identical with the standard Keldysh amplitude, Eq. (2.8). That is, we have for the three-dimensional δ-function potential

$$M_{\mathrm{p,BT}}^{(0)}=M_{\mathrm{p}}^{(K)}\ , \tag{4.11}$$

and for the particular case of circular polarization,

$$M_{\mathrm{p}}^{(0)}=M_{\mathrm{p}}^{(K)}\ . \tag{4.12}$$

Specifically, for a circularly polarized monochromatic plane wave with vector potential

$$\mathbf{A}(t)=a(\hat{\mathbf{x}}\cos\omega t+\hat{\mathbf{y}}\sin\omega t)\ , \tag{4.13}$$

we have

$$M_{\mathrm{p}}^{(0)}=\frac{i\sqrt{\kappa}}{m}\sum_{n=-\infty}^{\infty}J_n\left[\frac{eap_T}{m\omega}\right]e^{in\delta}$$
$$\times\delta(E_{\mathrm{p}}+|E_0|+U_p-n\omega)\ , \tag{4.14}$$

where $p_T=\sqrt{p_x^2+p_y^2}$, $p_x=p_T\cos\delta$, $p_y=p_T\sin\delta$, and $U_p=e^2a^2/2m$.

The equality (4.12), even though it holds only in the special case of the three-dimensional δ-function potential and circular polarization, is quite remarkable. Recall that only the scattering term of the wave function (4.4) contributes to $M_{\mathrm{p}}^{(0)}$ while $M_{\mathrm{p}}^{(K)}$ derives solely from the plane-wave term.

The time integration in the first term of Eq. (4.8) cannot be carried out analytically. Fortunately, in many situations this term is small compared with the second term. A rough estimate suggests that the first term is of relative order

$$\frac{e^2\mathbf{A}\cdot\mathbf{E}}{(\kappa+ip)^3}\frac{m}{2\pi(\kappa-ip)}=\frac{\omega e^2\mathbf{A}^2m}{2\pi(\kappa^2+p^2)(\kappa+ip)^2} \tag{4.15}$$

with respect to the second. For $p\ll\kappa$, this ratio assumes the value $(e^2\mathbf{A}^2/2m\omega)(\omega/|E_0|)^2/(4\pi)$, which is quite small under the conditions where typical above-threshold-ionization experiments have been carried out. The term is, however, qualitatively important in that it violates the symmetry $\mathbf{p}\rightarrow-\mathbf{p}$, which is obeyed by the second term.

In order to calculate the first correction $M_{\mathrm{p}}^{(1)}$ we start from expression (3.20) and insert the eigenfunction expansion (2.3) for the propagator G_0. Due to inversion symmetry its bound-state part does not contribute. We obtain

$$M_{\mathrm{p}}^{(1)}=-\int dt\int^t dt'\Phi_{\mathrm{p}}(t')\int d^3p'\exp\left[i\frac{p^2-p'^2}{2m}t\right]\exp\left[i\left[\frac{p'^2}{2m}+|E_0|\right]t'\right]$$
$$\times\left\langle\psi_{\mathrm{p}}^{(0)}(\mathbf{r})\left|-\frac{e}{m}(\hat{\mathbf{p}}-\mathbf{p})\cdot\mathbf{A}(t)e^{ier\cdot\mathbf{A}(t)}\right|\psi_{\mathrm{p}'}^{(0)}(\mathbf{r})\right\rangle\langle\psi_{\mathrm{p}'}^{(0)}(\mathbf{r})|-e\mathbf{r}\cdot\mathbf{E}(t')|\psi_0^{(0)}(\mathbf{r})\rangle\ . \tag{4.16}$$

The two matrix elements can be readily evaluated. Obviously, the second matrix element is proportional to $\mathbf{p}'\cdot\mathbf{E}(t')f(\mathbf{p}'^2)$. As for the first, integrating by parts with respect to $\hat{\mathbf{p}}=-i\nabla$, one can convince oneself that it may depend on $\mathbf{A}^2$, $\mathbf{p}'\cdot\mathbf{A}$, $\mathbf{p}\cdot\mathbf{A}$, and $\mathbf{p}'^2$, but not on $\mathbf{p}'\cdot\mathbf{E}$ or on $\mathbf{p}\cdot\mathbf{p}'$. For circular polarization we have $\mathbf{A}\cdot\mathbf{E}=0$. Hence, upon the transformation $\mathbf{p}'\cdot\mathbf{E}(t')\rightarrow-\mathbf{p}'\cdot\mathbf{E}(t')$, we can conclude that

$$M_{\mathbf{p}}^{(1)}=0 \quad \text{(circ. pol.)} . \tag{4.17}$$

For any polarization other than circular, $M_{\mathbf{p}}^{(1)}$ is nonzero. The calculation of the higher-order terms $M_{\mathbf{p}}^{(n)}$ becomes increasingly cumbersome. We will be satisfied with a discussion of $M_{\mathbf{p}}^{(2)}$ for the circularly polarized field (4.13). We expect $M_{\mathbf{p}}^{(2)}$ to introduce, among other contributions, the ac Stark shift of the ground state

$$\Delta=\frac{m(ea\omega)^2}{8\kappa^4}=\frac{1}{16}\eta\omega\left[\frac{\omega}{|E_0|}\right]^2 . \tag{4.18}$$

Indeed, after a lengthy but straightforward calculation, we obtain to lowest order in

$$\eta=\frac{U_p}{\omega} \tag{4.19}$$

that

$$\sum_{n=0}^{2} M_{\mathbf{p}}^{(n)}=\frac{i\sqrt{\kappa}}{m}\sum_{n=-\infty}^{\infty} J_n\left[\frac{eap_T}{m\omega}\right]e^{in\delta} \times\delta(E_{\mathbf{p}}+|E_0|+U_p+\Delta-n\omega) \times\left[1+i\eta\left[\frac{\omega}{|E_0|}\right]^3 f(p,\kappa)\right] , \tag{4.20}$$

with

$$f(p,\kappa)=-\frac{1}{64\pi}(p^2+\kappa^2) \times\frac{25\kappa^3+29\kappa^2 ip-15\kappa p^2-3ip^3}{(\kappa+ip)^5} . \tag{4.21}$$

Hence the significance of corrections to the lowest-order result is governed by the parameter

$$V^2=2\eta\left[\frac{\omega}{|E_0|}\right]^3=2\frac{U_p}{|E_0|}\left[\frac{\omega}{|E_0|}\right]^2 . \tag{4.22}$$

The same parameter occurs in estimates of the multiphoton-ionization rate in intense fields in the asymptotic regime such that $\omega/|E_0|\ll V\ll 1$ (see Refs. [16,24]).

V. CONCLUSIONS

We have proposed a systematic expansion of the exact matrix element that governs multiphoton ionization of atoms in the presence of intense laser fields. Our method is based on a semiclassical description of the ionized electrons in the spirit of Keldysh's original approach. However, in contrast to the former, it fully allows for the role of intermediate states, and, in particular, of intermediate resonances. Hence it can be applied, e.g., to the calculation of resonantly enhanced multiphoton ionization. Because our expansion, as well as Keldysh's, is built on the Volkov solution as a vital ingredient, each term contains terms of arbitrarily high order in the atom-field interaction. Therefore each term displays an ATI series of peaks. The lowest-order term in the expansion yields the usual Keldysh approximation, expressed as in Keldysh's original paper [8] in the length gauge, provided the final state is replaced by a plane wave. The higher-order terms lead to successive approximations. The physical phenomena covered by these terms include transitions to excited states, resonances, and intensity-dependent level shifts. The optimal term in the expansion is likely to be given by the multiphoton order of the ionization process. For higher orders, the expansion is more and more replaced by ordinary perturbation theory in the atom-field interaction.

We hope that this method may allow for a more systematic study of the validity of Keldysh-like approximations, since in our case an exact prescription is given to calculate the successive corrections. We have exemplified this procedure with the three-dimensional δ-function potential, for which exact analytical expressions have been obtained not only for the first Keldysh-like term, but also for the first nonvanishing correction, thus allowing for the precise analytic characterization of the conditions under which the approximation is valid in this case. We also hope that the method may be useful for the calculation of resonantly enhanced multiphoton ionization, at least for not too high order. It can also conveniently be applied to the calculation of quantum corrections to free-free transitions.

Of course, our method suffers also from some of the limitations of the usual perturbation theory. Even though we are able to account, as in the conventional perturbation approach, for intermediate resonances through partial summations, our expansion should break down if the field becomes too intense. This fact does not preclude its usefulness, however, due to the explicit incorporation of the ATI peaks.

ACKNOWLEDGMENTS

One of us (W.B.) is grateful to M. Kleber and the theory group of the Munich Technical University for their hospitality and for stimulating discussions. W.B. is also indebted to V. Florescu and A. Cionga for pointing out Ref. [28] to him. This work was partially supported by the Office of Naval Research and the Sonderforschungsbereich SFB 338 of the Deutsche Forschungsgemeinschaft.

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