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Shortcomings of the Keldysh approximation

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A simple derivation of the Keldysh amplitude for the ionization of an atom by a strong field is presented using a perturbation expansion in the binding potential. It is shown that the "Keldysh approximation" is questionable under conditions of strong ionization, when there is substantial probability of removing the electron from its initial bound state. This is consistent with published numerical studies based on model binding potentials. It is argued that the Keldysh amplitude is not gauge invariant in the usual sense, and that it is effectively canceled in a conventional gauge-invariant formulation of strong-field perturbation theory.

The Keldysh theory of ionization by a strong oscillating field is characterized by the treatment of the detached electron as an otherwise free particle in the field.[1] The photoelectron is therefore described by a Volkov wave function, i.e., a solution of the Schrödinger equation for a charged particle in an external field.[2] Apart from its neglect of the Coulomb interaction of the electron with the residual ion, the Keldysh approximation is often regarded as a "nonperturbative" approach to strong-field atomic ionization.

In this paper the well-known form of the Keldysh transition amplitude involving an initial bound state and a final Volkov state is derived very simply using a perturbation expansion of the time evolution operator in the atomic binding potential $V(\mathbf{r})$. (As in much of the recent literature, the "Keldysh approximation" here refers to the lowest-order approximation to the formulation originally outlined by Keldysh.) In order to arrive at the Keldysh amplitude it is necessary to assume that the probability of the electron being removed from its initial state is small. The Keldysh approximation is therefore of questionable validity under conditions of strong ionization, when this probability is not small. This observation is consistent with *ab initio* numerical studies in which the Keldysh approximation has been found to be in serious disagreement, sometimes even qualitatively, with exact results.

Another difficulty with the Keldysh approximation is that it produces gauge-dependent results. This problem is discussed below in connection with a more conventional, *gauge-invariant* perturbation theory advocated by Antunes Neto and Davidovich.[3] This gauge-invariant approach is formally equivalent to that employed by Kroll and Watson[4] for the scattering of an electron in the presence of a strong field.

The Hamiltonian of interest has the form

$$H(t)=H_0(t)+V(\mathbf{r}) \tag{1a}$$

$$=p^2/2m+H_I(t)+V(\mathbf{r}) \tag{1b}$$

$$=H_A+H_I(t)\,, \tag{1c}$$

where $H_0(t)$ is the Hamiltonian for an electron in the applied field alone, V is the atomic binding potential, $H_I(t)$ is the interaction Hamiltonian for the coupling of the electron to the field [e.g., $H_I(t)=-e\mathbf{r}\cdot\mathbf{E}(t)$ or $H_I(t)=-(e/mc)\mathbf{A}(t)\cdot\mathbf{p}+(e^2/2mc^2)\mathbf{A}^2(t)$], and $H_A=p^2/2m+V$ is the unperturbed Hamiltonian for the bound electron in the absence of the applied field.

The transition amplitude of interest is

$$A_{fi}(t)=\langle f|U(t)|i\rangle\,, \tag{2}$$

where $|i\rangle$ is the initial (bound) state and $|f\rangle$ is a plane-wave state associated with a *free* electron of momentum $\mathbf{p}$. The time evolution operator $U(t)$, satisfying $i\partial U/\partial t=HU$, is given by

$$U(t)=U_0(t)u(t)\,, \tag{3}$$

where

$$i\partial U_0/\partial t=H_0U_0\,, \tag{4a}$$

$$i\partial u/\partial t=U_0^\dagger VU_0u=U_0^\dagger VU\,, \tag{4b}$$

and $U_0(0)=u(0)=1$. Thus

$$A_{fi}(t)=\langle f|U_0(t)|i\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')VU(t')|i\rangle\,, \tag{5}$$

$U(t')|i\rangle$ is the state to which $|i\rangle$ evolves after a time t'. If we assume that

$$U(t')|i\rangle\cong e^{iI_0t'}|i\rangle\equiv|\psi_i(t')\rangle\,, \tag{6}$$

where I_0 is the ionization potential associated with state $|i\rangle$, then

$$A_{fi}(t)\cong\langle f|U_0(t)|i\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')V|\psi_i(t')\rangle\,. \tag{7}$$

The approximation (6) assumes that the probability of the electron leaving its initial bound state is negligible, and also that the level shift due to the applied field may be ignored.

Now since

$$i\frac{\partial}{\partial t'}|\psi_i(t')\rangle=[p^2/2m+V]|\psi_i(t')\rangle\,, \tag{8}$$

we have

$$A_{fi}(t) \cong \langle f | U_0(t) | i \rangle - \int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') [i\partial/\partial t' - p^2/2m] | \psi_i(t') \rangle$$

$$= \langle f | \psi_i(t) \rangle + i \int_0^t dt' \langle f | U_0(t) [i\partial U_0^\dagger(t')/\partial t' + U_0^\dagger(t') p^2/2m] | \psi_i(t') \rangle , \tag{9}$$

after an integration by parts. But from (4a) and (1) it follows that

$$i\partial U_0^\dagger/\partial t' + U_0^\dagger(t') p^2/2m = -U_0^\dagger(t') H_I(t') , \tag{10}$$

and therefore that

$$A_{fi}(t) \cong \langle f | \psi_i(t) \rangle - i \int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') \; H_I(t') | \psi_i(t') \rangle . \tag{11}$$

The first term obviously remains bounded for all t and so does not contribute to a transition *rate* (defined as $|A_{fi}(t)|^2/t$ for $t \to \infty$). Thus the transition rate is determined by the amplitude

$$A_{fi}^K(t) = \int_0^t dt' \langle f | U_0^\dagger(t') H_I(t') | \psi_i(t') \rangle$$

$$= \int_0^t dt' \langle \psi_f^V(t') | H_I(t') | \psi_i(t') \rangle , \tag{12}$$

where we have dropped the factor $U_0(t)$ since in the end it only contributes to a physically irrelevant phase of the amplitude. The state

$$| \psi_f^V(t') \rangle = U_0(t') | f \rangle , \tag{13}$$

is the state to which an initial free-electron state evolves *under the action of the applied field alone.* That is, $| \psi_f^V(t') \rangle$ is a Volkov state, and (12) is in fact just the Keldysh approximation to the transition amplitude.

The crucial assumption in our derivation of the Keldysh amplitude is that the probability amplitude for the electron to remain in its initial state is close to unity. The approximation is therefore suspect under conditions of strong ionization, for instance. Indeed, several numerical experiments on simplified models have uncovered substantial differences between the predictions of the Keldysh approximation and the "exact" ionization probabilities determined numerically. For instance, Geltman[5] found that the standard "Oppenheimer" tunneling ionization formula, which can be obtained from the Keldysh approximation in the limit in which the adiabatic tunneling parameter γ is small, gave poor estimates of the ionization probability in a one-dimensional δ-function model for the atomic potential. More recently, Antunes Neto, Davidovich and Marchesin,[6] Javaneinen and Eberly,[7] and Collins and Merts[8] have found poor agreement between the Keldysh approximation and one-dimensional models under conditions where multiphoton absorption can be expected to be the dominant ionization mechanism.

It is interesting now to return to the *exact* expression (5) and employ a different expression for the (exact) time evolution operator U:

$$U(t) = \bar{U}_0(t) \bar{u}(t) , \tag{14a}$$

$$i\partial \bar{U}_0/\partial t = H_A \bar{U}_0 , \tag{14b}$$

$$i\partial \bar{u}/\partial t = \bar{U}_0^\dagger H_I \bar{U}_0 \bar{u} = \bar{U}_0^\dagger H_I U , \tag{14c}$$

and $\bar{U}_0(0) = \bar{u}(0) = 1$. This form, of course, is the basis for a perturbation expansion in the electron-field interaction H_I. Using it in (5) allows us to obtain the Keldysh amplitude plus a correction represented by the third term in the expression

$$A_{fi}(t) = \langle f | U_0(t) | i \rangle - i \int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') V | \psi_i(t') \rangle$$

$$- \int_0^t dt' \int_0^{t'} dt'' \langle f | U_0(t) U_0^\dagger(t') V \bar{U}_0(t') \bar{U}_0^\dagger(t'') H_I(t'') U(t'') | i \rangle , \tag{15}$$

where in the second term we have used the fact that $\bar{U}_0(t') | i \rangle = | \psi_i(t') \rangle$. Since the first two terms in (15) are identical to the right side of (7), we see from (11) that the exact amplitude (15) may be rewritten as

$$A_{fi}(t) = \langle f | \psi_i(t) \rangle - i \int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') H_I(t') | \psi_i(t') \rangle$$

$$- \int_0^t dt' \int_0^{t'} dt'' \langle f | U_0(t) U_0^\dagger(t') V \bar{U}_0(t') \bar{U}_0^\dagger(t'') H_I(t'') U(t'') | i \rangle . \tag{16}$$

The corrections to the Keldysh amplitude contained in the third term of (16) might appear at first glance to be small for strong fields because of the appearance of the small perturbation V to H_I. However, this is not the case. Note that

$$\bar{U}_0^\dagger H_I U = \bar{U}_0^\dagger (H - H_A) U = \bar{U}_0^\dagger (i\partial U/\partial t) + (i\partial \bar{U}_0^\dagger/\partial t) U = i\partial (\bar{U}_0^\dagger U)/\partial t , \tag{17}$$

and therefore

$$A_{fi}(t)=\langle f|\psi_i(t)\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')H_I(t')|\psi_i(t')\rangle$$
$$-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')VU(t')|i\rangle+i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')V\bar{U}_0(t')|i\rangle$$
$$=\langle f|\psi_i(t)\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')H_I(t')|\psi_i(t')\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')VU(t')|i\rangle-\langle f|\psi_i(t)\rangle$$
$$+\langle f|U_0(t)|i\rangle+i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')H_I(t')|\psi_i(t')\rangle$$
$$=\langle f|U_0(t)|i\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')VU(t')|i\rangle, \tag{18}$$

which is Eq. (5). This form, of course, suggests the conventional sort of perturbation expansion in which the form (4) of the time evolution operator is used exclusively:

$$A_{fi}(t)=\langle f|U_0(t)|i\rangle-i\int_0^t dt'\langle f|U_0(t)U_0^\dagger(t')VU_0(t')|i\rangle$$
$$-\int_0^t dt'\int_0^{t'} dt''\langle f|U_0(t)U_0^\dagger(t')VU_0(t')U_0^\dagger(t'')VU_0(t'')|i\rangle+\cdots. \tag{19}$$

In this perturbation expansion of the evolution operator the Keldysh amplitude does not appear. It has been canceled by the third term in (16). Such a cancelation occurs also in the Green-function analysis of Antunes Neto and Davidovich.[3] These authors, and more recently Mittleman,[9] showed that the Keldysh amplitude is not invariant under a contact transformation in which the $\mathbf{A}^2$ term is removed from the Hamiltonian, and they remarked that there are similar difficulties with gauge transformations. Let us take up briefly the question of gauge invariance in the present formulation.

Consider a gauge function $\chi(\mathbf{r},t)$ that vanishes at the initial and final times, $t=0$ and $t=T$, of interest. This could represent, for instance, the transformation from the $\mathbf{A}\cdot\mathbf{p}$ to the $\mathbf{r}\cdot\mathbf{E}$ form of the Hamiltonian, with the vanishing of χ at initial and final times corresponding to the adiabatic switching on and off of the vector potential.[10] Without the gauge transformation the transition amplitude is $A_{fi}(T)=\langle f|U(T)|i\rangle$. Under the gauge transformation $|\psi\rangle\rightarrow|\psi'\rangle=e^{-i\chi}|\psi\rangle$ the evolution operator $U\rightarrow U'=e^{-i\chi}U$, and the transition amplitude becomes

$$A'_{fi}(T)=\langle f|U'(T)|i\rangle=\langle f|e^{-i\chi(\mathbf{r},T)}U(T)|i\rangle=A_{fi}(T), \tag{20}$$

because of the assumption $\chi(\mathbf{r},0)=\chi(\mathbf{r},T)=0$. This gauge invariance of the exact transition amplitude also holds order by order in perturbation expansion (19) since, for instance,

$$\langle f|U_0(T)U_0^\dagger(t')VU_0(t')|i\rangle\rightarrow\langle f|U_0'(T)U_0'^\dagger(t')VU_0'(t')|i\rangle$$
$$=\langle f|e^{-i\chi(\mathbf{r},T)}U_0(T)U_0^\dagger(t')e^{i\chi(\mathbf{r},t')}Ve^{-i\chi(\mathbf{r},t')}U_0(t')|i\rangle$$
$$=\langle f|U_0(T)U_0^\dagger(t')VU_0(t')|i\rangle. \tag{21}$$

However, the Keldysh amplitude does not have this gauge invariance. Consider the effect of a gauge transformation on the second term in (7):

$$-i\int_0^T dt'\langle f|U_0(T)U_0^\dagger(t')V|i\rangle e^{iI_0t'}\rightarrow-i\int_0^T dt'\langle f|U_0'(T)U_0'^\dagger(t')V|i\rangle e^{iI_0t'}$$
$$=-i\int_0^T dt'\langle f|U_0(T)U_0^\dagger(t')e^{i\chi(\mathbf{r},t')}V|i\rangle e^{iI_0t'}$$
$$\neq-i\int_0^T dt'\langle f|U_0(T)U_0^\dagger(t')V|i\rangle e^{iI_0t'}. \tag{22}$$

This lack of gauge invariance explains why different results have been obtained when the Keldysh approximation has been used with the different interaction Hamiltonians $H_I=-e\mathbf{r}\cdot\mathbf{E}$ and $H_I=-(e/mc)\mathbf{A}\cdot\mathbf{p}+(e^2/2mc^2)\mathbf{A}^2$, which are connected by a gauge transformation with $\chi=e\mathbf{r}\cdot\mathbf{A}/\hbar$. For weak fields the Keldysh approximation essentially reproduces the results of conventional, first-order gauge-invariant perturbation theory in which H_I is treated as a small perturbation. This is evident from Eq. (12).

Thus the Keldysh amplitude in its most frequently quoted form is not gauge invariant. The more conventional perturbation theory based on (19), by contrast, is gauge invariant. Formally, this is precisely the kind of expansion implicit in the Kroll-Watson theory of electron scattering in a strong field. Antunes Neto and coworkers[3,6] have found in their model calculations using a δ-function potential that for strong fields the first term in (19) provides a much better approximation to the exact amplitude than the Keldysh approximation, which failed dramatically for the cases they presented. Based on the formal but simple approach used in this paper, it seems clear that the Keldysh approximation is likely to fail for strong fields. It does not, as sometimes claimed, provide a

"nonperturbative" description of strong-field ionization.

This does not contradict the fact that the Keldysh approximation accounts for things like the multiphoton peaks in the photoelectron energy distributions measured in above-threshold ionization. Virtually *any* approach that includes multiphoton absorption can be expected to predict, at least qualitatively, such structure. At the same time it must be recognized that the Keldysh theory appears to be in reasonably good quantitative accord with at least some above-threshold ionization experiments,[11] and remains a valuable benchmark in the theory of strong-field interactions.

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[1]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)]; H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[2]D. M. Volkov, Z. Phys. **94**, 250 (1935). Volkov solved the Dirac equation for an electron in an applied field, including the possibility that the field could be polychromatic.

[3]H. S. Antunes Neto and L. Davidovich, Phys. Rev. Lett. **53**, 2238 (1984).

[4]N. M. Kroll and K. M. Watson, Phys. Rev. A **8**, 804 (1973).

[5]S. Geltman, J. Phys. B **10**, 831 (1977).

[6]H. S. Antunes Neto, L. Davidovich, and D. Marchesin, in *Coherence and Quantum Optics V,* edited by L. Mandel and E. Wolf (Plenum, New York, 1985).

[7]J. Javaneinen and J. H. Eberly, J. Phys. B **21**, L93 (1988).

[8]L. A. Collins and A. L. Merts, Phys. Rev. A **37**, 2415 (1988).

[9]M. H. Mittleman (unpublished).

[10]In general, of course, the initial- and final-state vectors must be transformed to ensure gauge invariance. Under our assumption that the gauge function χ vanishes at the initial and final times, however, we can effectively ignore any transformation of $|i\rangle$ and $|f\rangle$.

[11]H. R. Reiss, J. Phys. B **20**, L79 (1987); J. Opt. Soc. Am. B **4**, 726 (1987).