Multiphoton-ionization transition amplitudes and the Keldysh approximation

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The Keldysh approximation to treat the multiphoton ionization of atoms is reconsidered. It is shown that, if one consistently uses the hypothesis under which the approximation should be valid (essentially, that of a weak, short-range binding potential), a Keldysh-like term results as an approximation to the first term of a uniformly convergent series in powers of the binding potential. No cancellation occurs when higher-order terms are taken into account. This result allows one to consider the Keldysh approximation as a well-defined theoretical model, without implying, however, that it is adequate to describe multiphoton ionization of real atoms.

The advent of very powerful lasers has made the experimental phenomenology of multiphoton ionization of atoms much richer. At the same time, it has become apparent that the lowest-order perturbation theory is unable, as a rule, to account satisfactorily for the latest experimental observations. The consequence is that at present there is no working theory able to account rigorously for the many features characteristic of multiphoton ionization of atoms, although several partially successful theoretical treatments have been devised and are available in the literature.[1]

In this context, a significant revival of interest has been enjoyed lately[2] also by the treatment originally devised by Keldysh in 1964 to treat the ionization of an electron by a strong electromagnetic field, bound by a short-range potential.[3] The Keldysh approximation (KA) was originally proposed as an ansatz rather than as a first term of some rigorously constructed expansion, and at the time raised considerable interest. In particular, an effort was made by several authors, especially in the Soviet Union, to clarify the physical contents of the model and to improve it. As a rule, the outcome of such other investigations was that, within the same physical assumptions (first of all, that of a short-range potential), but following different approaches, largely the same results were found.[4] In more recent years, there has been the effort to justify the KA as the first-order term of an expansion in the binding potential in the presence of a strong radiation field.[5]

The KA misses the information on the discrete atomic spectrum as well as of the final-state interaction between the ejected electron and the residual ion. Intuitively, a justification to it may be based on the Fermi golden rule; besides, it may be thought of as the strong-field generalization of the well-known plane-wave treatment of the conventional photoelectric effect, which for ionization far from threshold is known to perform satisfactorily. Thus the KA appears to be a model of immediate physical interpretation. Appealing features of the KA are its simple structure and easy handling; moreover, it has been found to reproduce in a qualitatively satisfactory way a number of features of the measured strong-field ionization.[6]

On the other hand, serious limits of the KA as well have been pointed out too.[7] In short, according to many different contributions and statements, the most likely conclusion on the KA might be that it is hardly adequate to describe multiphoton ionization of real atoms, but that at the same time it is of some use in getting preliminary information on some aspects of the process.

However, recently the reliability of the KA as a physically well-defined theoretical model has been seriously questioned by different authors.[8,9] These authors perform a perturbation expansion of the transition amplitude in the binding potential, and name after Keldysh the first term of the expansion. Considering higher-order terms of such an expansion, they show that the "Keldysh" term cancels. Thus, according to these analyses the KA appears to be merely a kind of artifact, in spite of its simple physical interpretation and of its satisfactory performance in a number of cases. While there is little doubt that the KA is generally inadequate, it is difficult to consider it simply as an artifact. Below, we show in fact that it is not the case, if one adheres to the original meaning of the Keldysh model. Our goal is obtained in the framework of the expansion suggested in Refs. 8 and 9. Nevertheless, we would like to point to the arbitrariness of calling after Keldysh the first term of an expansion in the binding potential V as done in Refs. 8 and 9.

We show first when the cancellation of the Keldysh term occurs and why; then, we show that the term named after Keldysh in the Refs. 8 and 9 is not fully consistent with the assumption of treating a weak, short-range binding potential; finally, we derive a transition amplitude consistent with such an assumption. The expansion of this latter transition amplitude in powers of the binding potential and a further weak-potential approximation produces a new transition amplitude having just the Keldysh-like structure. The next terms of the exact expansion do not cancel the first one and reproduce the uniformly convergent expansion suggested in Refs. 8 and 9.

As usual in ionization processes,[10] we use the advanced form of the transition amplitude

$$A_{fi}(t)=\langle f|U^{\dagger}(t)|i\rangle \ , \tag{1}$$

where $|f\rangle$ is a field-free plane wave and $|i\rangle$ the initial bound state. $U(t)$ lets $|f\rangle$ evolve from $t=0$ [with $U(0)=1$] to t, where it is a fully interacting (with both the field and the binding potential) state. As in the conventional approaches, we use two partitions of the full Hamiltonian $H(t)=p^2/2m+H_F(t)+V$, namely,

$$H(t)=H_A+H_F(t) \tag{2a}$$

and

$$H(t)=H_0(t)+V \ , \tag{2b}$$

where $H_A=p^2/2m+V$ drives the field-free electron-binding potential system and $H_0(t)$ the field-interacting otherwise free electron. $H_F(t)$ is the electron-field interaction.

The full evolution operator $U(t)$ obeys $i\hbar\partial U/\partial t=H(t)U(t)$; looking for a solution of the form

$$U(t)=U_A(t)u_F(t) \ , \tag{3}$$

where $i\hbar\partial U_A/\partial t=H_A U_A(t)$ with $U_A(0)=1$, one finds the integral equation

$$U(t)=U_A(t)\left[1-\frac{i}{\hbar}\int_0^t dt' U_A^{\dagger}(t')H_F(t')U(t')\right] . \tag{4}$$

Alternatively we may look for a solution as

$$U(t)=U_F(t)u_A(t) \ , \tag{5}$$

where $i\hbar\partial U_F/\partial t=H_0 U_F(t)$ and $U_F(0)=1$, obtaining

$$U(t)=U_F(t)\left[1-\frac{i}{\hbar}\int_0^t dt' U_F^{\dagger}(t')VU(t')\right] . \tag{6}$$

Using now the form (4) in (1) we have

$$A_{fi}=\langle f|U_A^{\dagger}(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U^{\dagger}(t')H_F(t')U_A(t')U_A^{\dagger}(t)|i\rangle \ . \tag{7}$$

Iterative use of (4) will now reproduce the conventional perturbative expansion in the electron-field interaction H_F. Using instead Eq. (6) in (1) we have

$$A_{fi}=\langle f|U_F^{\dagger}(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U^{\dagger}(t')VU_F(t')U_F^{\dagger}(t)|i\rangle \tag{8}$$

and the iterative use of (6) will yield the perturbative expansion in the binding potential V. This is the expansion proposed in Ref. 8.

Without approximations, we may use the form (6) of $U(t)$ in (7), obtaining

$$A_{fi}=\langle f|U_A^{\dagger}(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^{\dagger}(t')H_F(t')U_A(t')U_A^{\dagger}(t)|i\rangle$$
$$+\left[\frac{i}{\hbar}\right]^2\int_0^t dt'\int_0^{t'} dt''\langle f|U^{\dagger}(t'')VU_F(t'')U_F^{\dagger}(t')H_F(t')U_A(t')U_A^{\dagger}(t)|i\rangle \ . \tag{9}$$

According to Refs. 8 and 9 the Keldysh approximation corresponds to retaining only the first row of the amplitude (9), namely, in describing the ionization as a single-step process from a bound state (undressed by the field) to a Volkov wave (i.e., a free electron interacting only with the field); thus, this approximation amounts to neglect the binding potential V which appears in the second row; the difficulty with this procedure and its interpretation is probably that the binding potential V is present to all orders in the atomic evolution operator $U_A(t')$ entering the Keldysh term, and there left unchanged; things are not improved attempting an expansion in V by approximating $U(t')$ by $U_F(t')$ in the second row of (9); again, this procedure would be incomplete, because $U_A(t')$ is left unchanged elsewhere. This feature leads to what follows. Using

$$U_F^{\dagger}(t)H_F(t)=-i\hbar\partial U_F^{\dagger}/\partial t-U_F^{\dagger}(p^2/2m) \tag{10}$$

by means of an integration by parts the Keldysh term transforms as

$$\langle f|U_A^{\dagger}(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^{\dagger}(t')H_F(t')U_A(t')U_A^{\dagger}(t)|i\rangle=\langle f|U_F^{\dagger}(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^{\dagger}(t')VU_A(t')U_A^{\dagger}(t)|i\rangle \tag{11}$$

and

$$\left[\frac{i}{\hbar}\right]^2\int_0^t dt'\int_0^{t'} dt''\langle f|U^{\dagger}(t'')VU_F(t'')U_F^{\dagger}(t')H_F(t')U_A(t')U_A^{\dagger}(t)|i\rangle$$
$$=\frac{i}{\hbar}\left[-\int_0^t dt'\langle f|U^{\dagger}(t')VU_A(t')U_A^{\dagger}(t)|i\rangle+\int_0^t dt'\langle f|U^{\dagger}(t')VU_F(t')U_F^{\dagger}(t)|i\rangle\right]$$
$$+\left[\frac{i}{\hbar}\right]^2\int_0^t dt'\int_0^{t'} dt''\langle f|U^{\dagger}(t'')VU_F(t'')U_F^{\dagger}(t')VU_A(t')U_A^{\dagger}(t)|i\rangle \tag{12}$$

so that if one now approximates $U(t)$ by $U_F(t)$ here, the first term cancels the time integral of the Keldysh term (11).

In (8), as no $U_A(t)$ appears, a perturbative procedure with respect to the binding potential may actually be done, just replacing $U(t)$ by $U_F(t)$. The up-to-first-order term in V of that expansion

$$A_{fi}^{(1)}=\langle f|U_F^\dagger(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^\dagger(t')VU_F(t')U_F^\dagger(t)|i\rangle \tag{13}$$

reads as a bound state dressed by the field that, interacting with the binding potential, ionizes to a Volkov state. This is the procedure suggested in Refs. 8 and 9 for weak binding potentials and strong fields.

Let us look for the following form of the full evolution operator:

$$U(t)=U_0(t)u_0(t)\ , \tag{14}$$

where $U_0(t)$ is the plane-wave evolution operator, obeying $i\hbar\partial U_0/\partial t=(p^2/2m)U_0(t)$. We find in this case the integral equation

$$U(t)=U_0(t)\left[1-\frac{i}{\hbar}\int_0^t dt'U_0^\dagger(t')[H_F(t')+V]U(t')\right] \tag{15}$$

yielding for the transition amplitude (1)

$$A_{fi}=\langle f|U_0^\dagger(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U^\dagger(t)[H_F(t')+V]U_0(t')U_0^\dagger(t)|i\rangle\ . \tag{16}$$

The zeroth order in V of the amplitude (16), using (6), is

$$A_{fi}^{(0)}=\langle f|U_0^\dagger(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^\dagger(t')H_F(t')U_0(t')U_0^\dagger(t)|i\rangle\ . \tag{17}$$

Equation (17) is exact, as long as the zeroth-order in V is concerned. If the initial state is very weakly bound, one can make the simplification $U_0(t)|i\rangle\cong\exp(iI_0t/\hbar)|i\rangle$, I_0 being the ionization energy of the bound state, obtaining

$$A_{fi}^{(0)}=\langle f|U_0^\dagger(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^\dagger(t')H_F(t')U_0^\dagger(t)\exp(iI_0t'/\hbar)|i\rangle\ , \tag{18}$$

which has the structure of the Keldysh term. For weak radiation fields, $U_F(t)\cong U_0(t)$ and Eq. (18) reproduces the transition amplitude for the plane-wave photoelectric effect. Of course, as Eq. (18) is an approximation to the gauge-invariant expression (17),[8,9] some gauge dependence is generally to be expected.

Let us now come back to the exact amplitude (16), to establish its relationship with the expansion in V already proposed in Refs. 8 and 9. From (16) and (6) the first-order term in V reads

$$\overline{A}_{fi}^{(1)}=\left[\frac{i}{\hbar}\right]^2\int_0^t dt'\int_0^{t'}dt''\langle f|U_F^\dagger(t'')VU_F(t'')U_F^\dagger(t')H_F(t')U_0(t')U_0^\dagger(t)|i\rangle+\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^\dagger(t')VU_0(t')U_0^\dagger(t)|i\rangle\ . \tag{19}$$

Using now Eq. (10), an integration by parts on the double-time integral in (19) yields

$$\overline{A}_{fi}^{(1)}=\frac{i}{\hbar}\int_0^t dt'\langle f|U_F^\dagger(t')VU_F(t')U_F^\dagger(t)|i\rangle\ , \tag{20}$$

which is exactly the time integral entering Eq. (13), namely, the first-order term of the expansion procedure in V already found in Refs. 8 and 9; of course, this should have been expected, because of the uniqueness of the expansion in power series. Analogously, use of (10) in (17) transforms this latter one in $\langle f|U_F^\dagger(t)|i\rangle$, namely, in the zeroth-order term in V of Eq. (13): as in Ref. 8 this term has been shown to be the leading one for strong fields, the meaning and purpose of the Keldysh ansatz is recovered. The way of getting Eq. (18) clarifies through which steps the Keldysh model arises and how the cancellation is removed; as discussed below an expansion in V is probably not particularly suited for ionization; nevertheless, accurate comparisons with different kinds of expansion (or models) based on real atom situations could prove very useful.

In summarizing, the Keldysh ansatz for treating the multiphoton ionization of atoms may be understood within the assumption of a weak short-range binding potential. Probably, the origin of the present controversy on the rating of the Keldysh approximation may be traced back to the choice by some authors to obtain the Keldysh amplitude by a formal derivation, based on a series expansion in which the binding potential is the perturbation. To see the difficulty of such a program, consider the case of an electron in the presence of a binding potential V and a radiation field $\mathbf{A}(t)$. If one is interested in the ionization process, the most natural way of proceeding is to consider the radiation field as the perturbation. The physical picture is then that of an initially bound electron evolving towards a free state under the influence of two fields. In an ionization process one has the peculiar situation that the binding potential is necessarily dominating over the radiation field in the initial state, while the reverse may be true in the final state. Thus, for ionization, a formal and consistent expansion in terms of V is expected to be not particularly transparent (as it is instead for scattering processes), although of course it can be done. In fact, one has the problem of the identification of the true first term. In particular, as clearly realized by Geltman and Teague,[11] who first adopted such an approach, care must be exercised in choosing the appropriate transition amplitudes.

When the expansion in V is done, one is in practice adopting a scattering theory picture, where a free electron embedded initially in the radiation field is scattered (multiply) by the binding potential V again into a free-state embedded in the field. No wonder then that one arrives at a series which has the formal structure of the charged particle scattering by V in the presence of a strong radiation field. To account for the fact that initially the electron is bound instead of being free one has to provide the free electron in the initial state with the distribution of momenta it has in the bound state. The original Keldysh amplitude is instead physically simple, transparent, and direct, and may be accepted on the assumption of a weak, short-range binding potential. Of course it puts obvious limitations on the range of validity of the KA, and on its ability to serve as a tool for interpreting the experimental phenomenology of real atoms. It could be the end of the story. Further, if one wants to put the Ka on a more formal basis, one can refer to the above derivation showing that the Keldysh term actually results as a weak, short-range potential approximation to the first term of a gauge-invariant and convergent expansion in series of the binding potential, whose next terms not only do not cancel the first one but reproduce the series usually reported in literature when treating the regime of strong radiation fields and weak binding potentials.

We believe that the above discussion should remove the suspicion that the time-honored Keldysh approximation is a physically ungrounded ansatz; instead, using the words of Ref. 9, it "remains a valuable benchmark in the theory of strong-field interactions"; at the same time, we maintain that it is generally inadequate to describe multiphoton ionization of real atoms, because of the many simplifications inherent in it.

A work has appeared recently[12] in which the analysis of the Keldysh approximation is carried on in detail with the conclusion to regard it as an ansatz rather than as the leading term in a perturbation series, in substantial agreement with our standpoint.

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