Convergence of High-Intensity Expansions for Atomic Ionization

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We show that a frequently used nonperturbative approximation for atomic ionization rates is canceled out when corrections are taken into account. This explains the strong gauge dependence of previous results. A convergent and gauge-invariant expansion is obtained. Numerical results show that its first term, which may be calculated analytically in many cases, describes very well the time-dependent behavior of the ionization probability for very strong fields.

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The interaction of strong electromagnetic fields with atoms and molecules has attracted much interest from both theoretical and experimental points of view. Perturbation theory has been extensively validated by experiments. Nonperturbative treatments have been very successful for scattering problems, where good agreement has been obtained between theory and experiment.[1,2] On the other hand, the situation looks much bleaker for multiphoton ionization. While high-order perturbation theory has been successfully compared with experiment,[3] for intensities up to 10^{15} W/cm^2, nonperturbative treatments, stemming from the pioneer work of Keldysh,[4] are confronted with several difficulties. Although one claim of experimental verification has been presented,[5] these theories do not lead to gauge-invariant transition rates, and their range of validity has remained obscure. Removal from the nonrelativistic Hamiltonian of the term proportional to the square of the vector potential $\vec{A}$, which corresponds to a contact transformation and leaves the exact transition rate invariant, drastically changes the outcome of the calculations. Numerical tests have been in sharp disagreement with Keldysh's result.[6,7]

Keldysh obtains the transition rate from the following *Ansatz* for the transition amplitude:

$$T_{fi}=\int_0^t dt' \langle \psi_f | H_I(t') \phi_i \rangle , \tag{1}$$

where H_I is the interaction between the atom and the electromagnetic field (in dipole approximation), $\phi_i(\vec{x},t)=\phi_i(\vec{x})\exp(iI_0t)$ is the initial bound state with energy $-I_0$ (in atomic units), and $\psi_f(\vec{x},t)$ is a plane-wave solution of the Schrödinger equation for an electron interacting with a plane electromagnetic wave. For the Coulomb-gauge interaction

$$H_I(\vec{x},t)=\vec{p}\cdot\vec{A}(t)/c+\vec{A}(t)^2/2c^2, \tag{2}$$

we have

$$\psi_f(\vec{x},t) = (2\pi)^{-3/2}\exp\{ipx - i\int_0^t dt'[\vec{p}+\vec{A}(t')/c]^2/2\}.$$

According to Keldysh, this state takes into account the main effect of the applied field, namely, the acceleration of the ionized electron.

Brandi, Davidovich, and Zagury[8] have shown how this *Ansatz* can be obtained from conventional Green's-function theory: It actually corresponds to the first term of an expansion of the exact Green's function in terms of the intra-atomic potential. Precisely the same kind of expansion is involved in other nonperturbative treatments.[9]

In this Letter, we show that higher-order terms in that series actually cancel out the contribution from (1) to the transition rate. After the cancellation is carried out, one gets another expansion in the intra-atomic potential, leading to gauge-invariant transition rates, which are also unaltered by removal of the A^2 term. Convergence of this series is rigorously proven for all values of the electromagnetic intensity and for suitable intra-atomic potentials. Furthermore, we derive analytical expressions for the first term of the expansion, which accounts for the major contribution for very strong fields and admits a simply physical interpretation.

We consider, for simplicity, a nonrelativistic one-electron atom, neglecting the motion of the nucleus. The interaction of this atom with the electric field $\vec{E}(t)$ is described by the Hamiltonian $H=H_0+H_I$, where $H_0=\vec{p}^2/2+V(\vec{r})$ and H_I is

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given by (2) with $\vec{A}(t) = -c\int_0^t dt'\vec{E}(t')$.

Let G, G_0, and G' be the retarded Green's functions associated with H, H_0, and $H' = p^2/2 + H_I$, respectively. These functions are related by (symbolically)

$$G = G_0 + GH_I G_0, \tag{3}$$

$$G = G' + GVG'. \tag{4}$$

Let the atom be, at the initial time $t=0$, in the bound state $|\phi_i\rangle$, which is an eigenstate of H_0. After a time t, the probability amplitude of finding the system in an eigenstate $|\phi_f\rangle$ of H_0 corresponding to the eigenvalue E_f is

$$A_{fi}(t) = i\langle\phi_f|G(t,0)\phi_i\rangle. \tag{5}$$

The ionization probability at time t is $P(t) = \sum_f |A_{fi}|^2$, the sum being carried out over all positive-energy eigenstates.

Using Eq. (3), we may rewrite (5) as

$$A_{fi} = \delta_{fi} + \int_0^t dt'\langle\phi_f|G(t,t')H_I\phi_i\rangle. \tag{6}$$

From (4) and (6), we get $A_{fi} = A_{fi}^K + A_{fi}^R$, where

$$A_{fi}^K = \delta_{fi} + \int_0^t dt'\langle\phi_f|G'(t,t')H_I\phi_i\rangle, \tag{7}$$

$$A_{fi}^R = \int_0^t dt''\langle\phi_f|G(t,t'')VG'(t'',t')H_I\phi_i\rangle. \tag{8}$$

Setting, in (7),

$$\phi_f \approx \phi_{\vec{p}} = (2\pi)^{-3/2}\exp(i\vec{p}\cdot\vec{x} - i\vec{p}^2 t/2)$$

yields precisely Keldysh's *Ansatz* except for an irrelevant phase factor.[8] Therefore, this *Ansatz* corresponds to two approximations in the exact transition amplitude (6): $G \approx G'$ and $\phi_f \approx \phi_{\vec{p}}$. Improvements of expression (1) may thus be obtained by correcting the final state (this is mandatory close to the ionization threshold) and considering higher-order terms in the expansion $G = G' + G'V \times G' + \ldots$. These higher-order terms are added up in A_{fi}^R.

In the following, we shall assume that ϕ_f in (7) is an eigenstate of H_0, and not of the momentum. We deal therefore with an "improved Keldysh *Ansatz*." Analogous procedures may be applied, however, to the original *Ansatz*, leading to similar conclusions.

Replacing $G'H_I$ in (7) by

$$(-i\partial/\partial t' + \nabla'^2/2)G'(x,x') - \delta^4(x-x'),$$

we get, after a partial integration with respect to x' $[x \equiv (\vec{x},t)]$, a decomposition of A_{fi}^K into two parts, $A_{fi}^K = A_{fi}^0 + A_{fi}'$, where

$$A_{fi}^0 = i\langle\phi_f|G'(t,0)\phi_i\rangle, \tag{9}$$

$$A_{fi}' = \int_0^t dt'\langle\phi_f|G'(t,t')V\phi_i\rangle. \tag{10}$$

The term A_{fi}^0 corresponds to the propagation of the electron wave packet under the sole action of the applied electric field. Since $G'(t,0) \propto t^{-3/2}$, this term does not contribute to the transition rate when $t \to \infty$. The relevant contribution comes therefore from A_{fi}'.

The invariance problems are manifested in Eq. (10). Upon removal of the A^2 term, H' gets transformed into $\bar{H}'$, to which there corresponds a Green's function

$$\bar{G}'(t,t') = G'(t,t')\exp\left[i\int_{t'}^t dt''\frac{\vec{A}^2}{2c^2}\right].$$

The same relation holds between the Green's functions G and $\bar{G}$ corresponding to H and $\bar{H} = H - \vec{A}(t)^2/2c^2$, respectively. Therefore, if $\bar{A}_{fi}$ is the transition amplitude corresponding to $\bar{H}$, calculated between the *same* eigenstates of H_0 as before, one has, from (5), $|\bar{A}_{fi}| = |A_{fi}|$, since the phase factor does not depend on space variables, and therefore can be taken out of the scalar product. We also have, from (9), $|\bar{A}_{fi}^0| = |A_{fi}^0|$. However, $|\bar{A}_{fi}'| \neq |A_{fi}'|$, since A_{fi}' involves an integration over time, and therefore different results will be obtained for the approximate transition rate, depending on whether H or $\bar{H}$ is used, in spite of the fact that the exact transition rate is invariant under this transformation. Analogous considerations hold for gauge transformations. If the gauge function $\chi(\vec{x},t)$ vanishes for the initial and final times, $|A_{fi}|$ and $|A_{fi}^0|$ remain invariant, while $|A_{fi}'|$ is modified. Of course, all of them change if $\chi(\vec{x},t) \neq 0$ or $\chi(\vec{x},0) \neq 0$, unless ϕ_i and ϕ_f are also transformed.

As in the case of A_{fi}^K, the term A_{fi}^R may also be decomposed, in the following way:

$$A_{fi}^R = -\int_0^t dt'\langle\phi_f|G(t,t')V\phi_i\rangle + i\int_0^t dt'\langle\phi_f|G(t,t')VG'(t',0)\phi_i\rangle + \int_0^t dt'\int_0^t dt''\langle\phi_f|G(t,t'')VG'(t'',t')V\phi_i\rangle. \tag{11}$$

Setting $G \approx G'$ in this expression, which amounts to considering the lowest-order correction to A_{fi}^K, we see that the first term on the right-hand side exactly cancels A_{fi}'. The third term on the right-hand side, which

also presents invariance problems, is canceled out when the next-order correction is considered. The cancellation to all orders of noninvariant terms can be seen by placing (4) into (11), so that

$$A_{fi}^{R}=-\int_0^t dt'\langle\phi_f|G'(t,t')V\phi_i\rangle+i\int_0^t dt'\langle\phi_f|G(t,t')VG'(t',0)\phi_i\rangle. \tag{12}$$

One cannot say, therefore, that A_{fi}^{R} is smaller in magnitude than A_{fi}^{K}. Adding up (9), (10), and (12), we get $A_{fi}(t)=\sum_{n=0}^{\infty}A_{fi}^{n}(t)$, where $A_{fi}^{n}(t)=(\phi_f,\psi^n)$, and $\psi^n=iG'(VG')^n\phi_i$. This expansion could be obtained directly from (4) and (5). It does not suffer from the invariance problems discussed above. Furthermore, its convergence can be established for any value of the applied field, as we will now show.

If ϕ_f is normalized to one (this involves taking a wave packet for the final state), then by Schwarz's inequality $|A_{fi}^{n}|\leq||\psi^n||$. Defining as usual the norm of an operator F by $||F||^2=\sup(F\psi,F\psi)/(\psi,\psi)$ for all $\psi\in L^2$, and using the fact that $||FK||\leq||F||\,||K||$ and $||G'||=1$, we get

$$||\psi^n||\leq\int_0^t dt_1\cdots\int_0^{t_{n-1}}dt_n||V||^n=t^n||V||n/n!,$$

so long as $||V||<\infty$. Therefore $\sum_n A_{fi}^{n}$ is absolutely convergent, if $V(\vec{x})$ is a bounded operator.

For one-dimensional problems, convergence can be established for absolutely integrable potentials, so long as the initial and final states are absolutely integrable. In this case, $|G'(t,t')|=\theta(t-t')[2\pi(t-t')]^{-1/2}$, so that $|A_{fi}^{n}|\leq a_n$, where

$$a_n=(2\pi)^{-(n+1)/2}\int dx|\phi_f(x,t)|\int dx'|\phi_i(x')|[\int dx|V(x)|]^n \times\int_0^t dt_1\cdots\int_0^{t_{n-1}}dt_n(t-t_1)^{-1/2}\cdots(t_{n-1}-t_n)^{-1/2}t_n^{-1/2}.$$

After a change of variables, the time integrals are decoupled, so that

$$a_{n+1}/a_n=(t/2\pi)^{1/2}\int dx'|V(x')|\int_0^1 dx\,x^{(n-1)/2}(1-x)^{-1/2}\approx(t/n)^{1/2}\int dx|V(x)|$$

for $n\gg 1$, and $\lim_{n\to\infty}a_{n+1}/a_n=0$, implying the absolute convergence of $\sum_n A_{fi}^{n}$.

We have obtained, therefore, an expansion which does not present the invariance problems of other nonperturbative approaches, and whose convergence can be rigorously established. Precisely the same expansion is involved in scattering problems.[1] Presumably, convergence will be faster for higher-intensity fields. In fact, for very strong fields, the main contribution should come from $A_{fi}^{0}=(\phi_f,\psi^0)$. Analytical expressions for ψ^0 and A_{fi}^{0} may be obtained in many cases: ψ^0 is the free-particle solution evolving from ϕ_i, centered around the classical position $X_{\rm cl}(t)=-\int_0^t dt'\int_0^{t'}dt''E(t'')$ of an electron initially at rest at the origin, moving under the action of the electric field $E(t)$. Thus, for a one-dimensional delta-function potential $V(x)=-B\delta(x)$, $B>0$, for which $\phi_i(x)=B^{1/2}\exp(-B|x|)$ and $E_i=-I_0=-B^2/2$, we get (the A^2 term is eliminated, since it contributes an irrelevant phase)

$$\psi^0(x,t)=\theta(t)B^{1/2}\{M(-x+X_{\rm cl}(t),-iB,t)+M(x-X_{\rm cl}(t),-iB,t)\}.$$

The function $M(x,k,t)$ has been introduced by Moshinsky,[10] and is given by

$$M(x,k,t)=\tfrac{1}{2}\exp[i(kx-k^2t/2)]\,\mathrm{erfc}[w\exp(-i\pi/4)],$$

where $w=(x-kt)/(2t)^{1/2}$ and $\mathrm{erfc}(z)=(2/\sqrt{\pi})\times\int_z^{\infty}\exp(-t^2)dt$. It is the solution of the free-particle Schrödinger equation which, for $t=0$, reduces to the cutoff exponential wave packet $M(x,k,0)=\theta(-x)\exp(ikx)$.

The time-dependent behavior of $A_{fi}^{0}(t)$ may thus be simply understood in terms of the classical motion of the particle and the quantum-mechanical spreading of the wave packet, since A_{fi}^{0} measures the overlap between the wave packet at time t and $\phi_f(x)$. This can be seen in Fig. 1, which displays the approximate ionization probability $P_0=1-|A_{ii}^{0}|^2$ [which may also be expressed in terms of $M(x,k,t)$] as a function of $\tau=I_0t$, for $E(t)=E_0\times\cos\omega t$. The spreading time of the initial wave packet is $t_s\approx I_0^{-1}$, so that, if T is the period of the oscillating field, $t_s/T\approx\omega/2\pi I_0$. On the other hand, the ratio η between the maximum value of $|X_{\rm cl}|$ and the initial width B^{-1} is $\eta\approx 8(E_0/E_c)\times(I_0/\omega)^2$, where $E_c=(2I_0)^{3/2}$ (for a hydrogen atom, E_c would be the internal electric field at the first Bohr orbit). These two ratios play an important role in the behavior of $P_0(\tau)$. If $I_0/\omega\lesssim(2\pi)^{-1}$, the wave packet spreads more slowly than it oscillates. Then, if $\eta\gtrsim 1$, $P_0(\tau)$ will

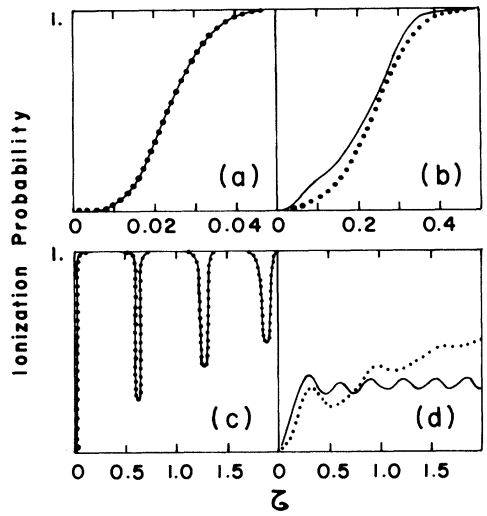

FIG. 1. Ionization probability as a function of $\tau = I_0 t$. Solid lines, exact results; dotted lines, $P_0(\tau)$. (a) $I_0/\omega = 20$, $E_0/E_c = 10^3$; (b) $I_0/\omega = 20$, $E_0/E_c = 10$; (c) $I_0/\omega = \frac{1}{10}$, $E_0/E_c = 10^3$; (d) $I_0/\omega = \frac{1}{10}$, $E_0/E_c = 10$.

have an oscillatory behavior [Fig. 1(c)], where $I_0/\omega = \frac{1}{10}$ and $\eta = 80$]. As η gets smaller than one, $X_{\rm cl}$ does not leave the region $|x| \leq B^{-1}$, and the amplitude of oscillation of $P_0(\tau)$ becomes smaller [Fig. 1(d), for which $\eta = 0.8$]. In both situations the ionization time t_I is given by the spreading time of the wave packet, that is, $t_I \approx I_0^{-1}$, or $\tau_I \approx 1$. In the limit $I_0/\omega >> (2\pi)^{-1}$, we have $t_s << T$, and $P_0(\tau)$ does not exhibit an oscillatory behavior. Then $\eta >> 1$ necessarily and t_I coincides with the time needed for the wave packet to leave the initial region: $t_I \approx t_s(E_c/E_0)^{1/2}$, or $\tau_I \approx (E_c/E_0)^{1/2}$. This is verified in Figs. 1(a) and 1(b).

The approximation $P_0(\tau)$ is compared in Fig. 1 with numerical solutions developed in Ref. 7 for the delta-function potential. Both the exact solution and $P_0(\tau)$ can be shown to depend only on E_0/E_c and I_0/ω. The approximation gets better as these two parameters increase, and converges to the numerical solution when $E_0/E_c >> 1$. We can also see from Fig. 1 that, for very strong fields, the evolution of the system would be poorly represented by a time-independent transition rate.

A similar analysis can be applied to other cases, taking, for instance, for ϕ_i hydrogenic states in three dimensions. A more detailed discussion will be presented elsewhere.

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