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Limits of a stationary phase method for ionization of atoms in strong laser fields

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Abstract

A stationary phase approach to a typical S -matrix expansion for atomic ionization in strong laser fields is examined with respect to its range of validity.

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1. Introduction

Ionization of an atom induced by an intense laser field and related processes, such as high harmonic generation and nonsequential double ionization, have been studied extensively during the past two decades. In the domain of nonperturbative electron–field interaction exact calculations of the process probability by direct numerical solution of the associated Schrödinger equation of the system of interest require large computational effort. Hence, systematic approximation methods, such as the S -matrix theory, are often used for an estimation of the transition amplitude. For example, one of the most well-known formula

for the total rate of ionization of atoms, the so-called Keldysh–Faisal–Reiss (KFR) formula [1], can be obtained as the leading term of a S -matrix expansion.

In the application of S -matrix theory to strong-field processes often Fourier-type integrals with large parameters in the exponent occur. Such integrals can asymptotically be reduced to the contribution of a few terms by means of the stationary phase method (e.g., [2,3]). The method has been used in the case of high harmonic generation [4], high-energy above threshold ionization [5–7] and nonsequential double ionization [8,9] to approximately evaluate terms of the respective S -matrix expansion.

Below we examine the range of validity of the stationary phase ansatz for a typical strong-field S -matrix expansion. To this end, we apply the stationary phase method to the calculation of the total rate of ionization of hydrogen atom within the first-order

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KFR theory. Then we compare the results obtained using this approximation with those of the exact evaluation of the KFR ionization rate.

2. Application of the stationary phase method

Using the standard KFR ansatz of first order for the wavefunction of an initially bound electron subjected to intense laser radiation, the transition amplitude to a Volkov state at time t can be written as (Hartree atomic units, $e = \hbar = m = 1$ are used, e.g., [10])

$$A_{fi}^{(1)}(t) = -i \int_{t_i}^t dt_1 \langle \phi_{\mathbf{k}}^V(t_1) | -\frac{\mathbf{p} \cdot \mathbf{A}(t_1)}{c} + \frac{A^2(t_1)}{2c^2} | \phi_0(t_1) \rangle, \quad (1)$$

where $\mathbf{A}(t)$ denotes the vector potential of the electron–field interaction in the dipole approximation, ϕ_0 is the initial state wavefunction and $\phi_{\mathbf{k}}^V$ is the Volkov wavefunction of momentum \mathbf{k} . We take the absolute square of the amplitude and sum over all final state momenta to obtain

$$P_{\text{ion}}^{(1)}(t) = \int_{t_i}^t dt_1 \int_{t_i}^t dt_2 \int d\mathbf{k} V(\mathbf{k}, t_1) V(\mathbf{k}, t_2) \times \exp(-iS(\mathbf{k}, t_1, t_2)) |\tilde{\phi}_0(\mathbf{k})|^2, \quad (2)$$

where $V(\mathbf{k}, t) = -\mathbf{k} \cdot \mathbf{A}(t)/c + A^2(t)/2c^2$ and $\tilde{\phi}_0(\mathbf{k})$ is the Fourier transform of the initial state wavefunction. The phase $S(\mathbf{k}, t_1, t_2)$ consists of the joint action of the Volkov electron in the velocity gauge and the ionization potential I_P :

$$S(\mathbf{k}, t_1, t_2) = \int_{t_1}^{t_2} dt' \left[\frac{1}{2} \left(\mathbf{k}(t') - \frac{\mathbf{A}(t')}{c} \right)^2 + I_P \right]. \quad (3)$$

For a linearly polarized laser field with polarization direction along the z -axis, we separate in Eq. (2) the \mathbf{k} -integration into a part along the z -axis and those perpendicular to it. At high laser intensities the vector potential is assumed to be large and the stationary phase method (e.g., [2,3]) is applied to evaluate the k_z -integral. There is one stationary point k_s for each

pair (t_1, t_2) ,

$$k_s(t_1, t_2) = \frac{1}{c\tau} \int_{t_1}^{t_2} dt' A(t'), \quad (4)$$

and we get

$$P_{\text{ion}}^{(1)}(t) \approx 2\pi^2 \int_{t_i}^t dt_1 \int_{t_i}^t dt_2 \frac{V(k_s, t_1) V(k_s, t_2)}{\sqrt{2\pi i \tau}} \times \exp(-iS(k_s, t_1, t_2)) I(k_{\perp}), \quad (5)$$

where

$$I(k_{\perp}) = \int_0^{\infty} d(k_{\perp}^2) |\tilde{\phi}_0(k_{\perp}, k_s)|^2 \exp\left(-i\frac{\tau}{2} k_{\perp}^2\right), \quad (6)$$

$\tau = t_2 - t_1$ and

$$S(k_s, t_1, t_2) = \left(I_P - \frac{k_s^2}{2}\right)\tau + \frac{1}{2c^2} \int_{t_1}^{t_2} dt' A^2(t'). \quad (7)$$

Using

$$\phi_0(\mathbf{r}) = 2\lambda^{3/2} \exp(-\lambda r) Y_{00}(\hat{r}), \quad \lambda = Z$$

for the ground state of hydrogenic atoms, we evaluate

$$I(k_{\perp}) = 64\pi\lambda^5 g_4(k_s, \tau)$$

with

$$g_4(k_s, \tau) = \int_0^{\infty} dx \frac{1}{(x + \beta)^4} e^{-i\mu x} = \frac{U(1, -2, i\mu\beta)}{\beta^3} = \exp(i\mu\beta) \frac{E_4(i\mu\beta)}{\beta^3}, \quad (8)$$

where $\beta = \lambda^2 + k_s^2 > 0$ and $\mu = \frac{\tau}{2}$. $U(1, -2, z)$ denotes the confluent hypergeometric function, which can be expressed for purely imaginary arguments by means of the fourth exponential integral function E_4 .

Using the result in Eq. (5), we get

$$P_{\text{ion}}^{(1)}(t) \approx 16\lambda^5 \int_{t_i}^t dt_1 \int_{t_i}^t dt_2 \frac{V(k_s, t_1) V(k_s, t_2) g_4(k_s, \tau)}{\sqrt{2\pi i \tau}} \times \exp(-iS(k_s, t_1, t_2))$$

$$\begin{aligned}
&= 32\lambda^5 \operatorname{Re} \left\{ \int_{t_i}^t dt_1 \int_{t_1}^t dt_2 \right. \\
&\quad \times \frac{V(k_s, t_1)V(k_s, t_2)g_4(k_s, \tau)}{\sqrt{2\pi i\tau}} \\
&\quad \left. \times \exp(-iS(k_s, t_1, t_2)) \right\}. \quad (9)
\end{aligned}$$

In the last step we have ordered the time integrations so that $\tau \geq 0$. The integrand contains an integrable $\tau^{-1/2}$ singularity and can be evaluated by direct numerical integration, since for small imaginary arguments $z = ix$ the confluent hypergeometric function takes the form $(1 - \frac{1}{2}ix)/3$. Please note, that a non-integrable $\tau^{-3/2}$ singularity occurs in the final expression, if in Eq. (2) the stationary phase method is applied to the \mathbf{k} -integration in all three dimensions. Usually the integrations over t_1 and t_2 are then performed by introducing a cutoff parameter [4]. As shown here, this additional approximation can be avoided in the case of linear polarization by performing the k_\perp -integration exactly and applying the stationary phase method to the k_z -integration only. We may further stress that, in fact, for linear polarization an asymptotically large parameter in the phase is present in the field direction only.

3. Results and discussion

Since we have taken into account the first order term of the S -matrix series only, $P_{\text{ion}}^{(1)}$ is not the total ionization probability, e.g., it is not restricted by unity. But, it is possible to determine a well-defined ionization rate, since $P_{\text{ion}}^{(1)}$ is found to depend linearly on the interaction time. This is exemplified in Fig. 1, where $P_{\text{ion}}^{(1)}(T_p)$ is shown as a function of the pulse length T_p , for an intense laser pulse at 800 nm and 1.053×10^{15} W/cm² having a \sin^2 -pulse shape.

Thus, the validity of the present stationary phase ansatz can be examined by evaluation of effective rates per pulse of length T_p , defined by [11]

$$\Gamma_{\text{eff}}^+ = -\frac{1}{T_p} \ln(1 - P_{\text{ion}}(T_p)) \quad (10)$$

using $P_{\text{ion}} \approx P_{\text{ion}}^{(1)}$ (for $P_{\text{ion}}^{(1)} \leq 1$).

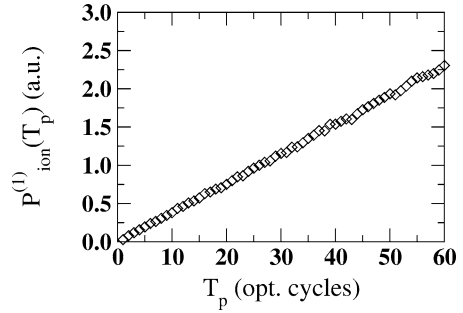


Fig. 1. $P_{\text{ion}}^{(1)}$ as a function of the pulse length. The laser parameters were 800 nm and 1.053×10^{15} W/cm², the temporal pulse shape was \sin^2 .

For the sake of comparison, we have calculated the exact KFR expression for the instantaneous ionization rate [1],

$$\begin{aligned}
\Gamma^+(I) &= 2\pi \sum_{N=N_0}^{\infty} \int d\hat{\mathbf{k}}_N k_N (U_p - N\omega)^2 \\
&\quad \times J_N^2\left(\alpha_0 \cdot \mathbf{k}_N; \frac{U_p}{2\omega}\right) |\tilde{\phi}_0(\mathbf{k}_N)|^2, \quad (11)
\end{aligned}$$

where $k_N^2/2 = N\omega - U_p - I_p$ is the kinetic energy of the electron after absorbing N photons, N_0 is the minimum photon number needed to be absorbed for ionization, $\alpha_0 = \sqrt{I}/\omega^2$ is the quiver radius and $U_p = I/4\omega^2$ is the ponderomotive energy of an electron in a laser field of intensity I and frequency ω . $J_N(a, b)$ is the generalized Bessel function of two arguments (e.g., [1c,9]). The exact rates are then used in the rate equation:

$$\frac{dP_{\text{ion}}(t)}{dt} = \Gamma^+(I(t))(1 - P_{\text{ion}}(t)), \quad (12)$$

which is solved under the constraint $P_{\text{ion}}(t=0) = 0$. The result is used in Eq. (10) to obtain the effective ionization rate for a finite pulse of length T_p .

In Fig. 2 we compare the effective rates as a function of the peak intensity obtained by using the stationary phase method (open diamonds) with the exact KFR results (lines) at different wavelengths between 400 and 1400 nm. The temporal profile of the pulse has been chosen to have a \sin^2 -shape of 10-cycle duration. It is seen from the figure that the results obtained using the stationary phase method agree well with the exact KFR rates, as expected, at high intensities. Below a critical intensity the approximative results are

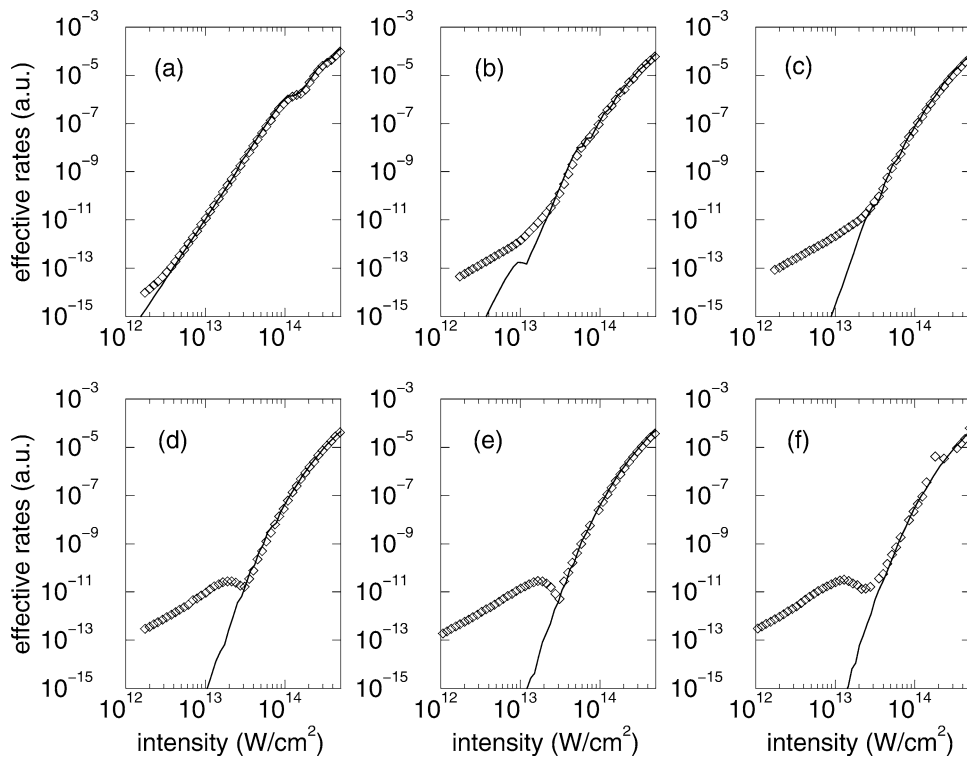


Fig. 2. Effective rates of ionization of the hydrogen atom by a strong laser pulse. Shown is a comparison between the results of a stationary phase approximation (open diamonds) to the first-order KFR theory and the exact KFR rates (lines). Calculations are performed for different wavelengths, namely (a) 400 nm, (b) 616 nm, (c) 800 nm, (d) 1064 nm, (e) 1200 nm, and (f) 1400 nm. A \sin^2 temporal pulse shape of 10-cycle duration is chosen.

found to systematically overestimate the exact effective rates. The results indicate that the application of the stationary phase method in a typical S -matrix expansion is justified as long as the intensity of the laser pulse is above mid of 10^{13} W/cm².

4. Summary

In conclusion, we have examined a stationary phase approximation to the first-order KFR theory of atomic ionization in a strong linearly polarized laser field. The stationary phase method has been applied to the momentum direction along the polarization direction of the field only, in order to avoid a typical non-integrable singularity in the final expression for the total ionization rate. It is found that the approximation is valid for intensities above mid of 10^{13} W/cm².

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