Tunneling ionization in a light field without Fourier expansion

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(Submitted 15 March 1995)
Zh. Éksp. Teor. Fiz. **108**, 456–463 (August 1995)

We find the transition amplitude for one version of the Keldysh–Faisal–Reiss model in the pA gauge by summing over periods of a laser pulse. Calculating the contribution of an individual period provides a simple, transparent picture of the quantum transition. Specifically, ionization in the tunneling regime can be viewed as a Landau–Zener transition from a bound state to a continuum state characterized by a definite momentum and time-dependent energy. The latter is equal to the kinetic energy of a classical electron in an electromagnetic field. We estimate the spectral width of photoelectrons in strong short laser pulses. © *1995 American Institute of Physics.*

1. INTRODUCTION

The model of ionization proposed by Keldysh[1] in 1964 has been actively exploited now for more than 30 years to treat the physics of the interaction of matter with strong electromagnetic fields. An attractive feature of the model is that it incorporates effects of a strong field without recourse to perturbation theory while retaining a simple analytic form. In a broad sense, the Keldysh model subsumes the theory in which a nonrelativistic Volkov solution is the final state in the quantum transition amplitude between a bound state and the continuum. In the Western literature, it is often called the Keldysh–Faisal–Reiss model. Its present status and its varied applications have been described in a number of recent publications.[2,3]

To a certain extent, the model's simplicity derives from the assumed constancy of the envelope of the oscillating laser field. This assumption makes it possible to expand the periodic factor in Fourier series and integrate over time in the expression for the transition amplitude. As a result, the amplitude ends up being proportional to an infinite sum of δ functions that express energy conservation for each harmonic component. Standard calculations then yield the transition rate (transition probability per unit time) per unit solid angle, which is equal to a sum of partial transition rates for the individual harmonic components.

Some remarks are in order concerning this computational scheme. In the tunneling limit, the many partial contributions are of comparable magnitude, and observables are determined by sums over a large number of harmonics. In general, the need to allow for a multitude of terms in a series suggests an inappropriate choice of expansion basis (a Fourier basis in the present instance). In the tunneling limit, an alternative picture of ionization, in which an electron tunnels through an oscillating potential barrier, provides a simpler, more transparent description.

The idea of an oscillating potential barrier can be introduced into the theory when the variable electric field can be described by a scalar potential. If we are using a vector potential, then no oscillating barrier enters directly into the calculations. Nevertheless, such a barrier is still referred to, serving as a qualitative explanation of the fact that in the tunneling limit, the ionization probability in a variable field is proportional to the penetrability of the static barrier.

The boundary of the continuum shifts by the mean value of the electron's oscillatory energy, due to energy conservation in the Fourier harmonics. The major role played by the shift in the formation of the photoelectron spectrum has been the subject of a great many multiphoton-based studies of above-threshold ionization.[2] In the tunneling limit, however, the continuum shift becomes enormous—much greater than the ionization potential of the free atom—and interpretation becomes difficult.

The effect of a variable field envelope on ionization is a problem of high interest, as current experiments involve laser pulses that are so strong and so short that the mean oscillatory energy varies by the full magnitude of the photon energy during a single optical period. Problems arise even at a lesser rate of field growth. For example, as long as the intensity (and thus the mean oscillatory energy) is fixed, energy conservation during a transition induced by the nth harmonic will relate the number of the harmonic to the energy of the photoelectron. As soon as we have a nonstationary field, uniqueness is lost: the electron has the same kinetic energy regardless of whether it is produced in photon channel n or $(n+1)$, as long as the mean oscillatory energy differs by the photon energy.[4] Similar problems arise when one considers nonlinear Compton scattering in a focused field.[5]

2. STATEMENT OF THE PROBLEM

The point of departure for our calculations is the set of standard expressions for the Keldysh–Faisal–Reiss (KFR) model in the pA gauge. The field strength F of a laser pulse of width τ is assumed to vary smoothly, and to vanish at infinite times in the past and future. According to the KFR model, throughout the time the field acts, the transition amplitude between the initial free-atom state ψ_i with potential I and the final free-electron state with momentum $\mathbf{p}$ and energy $\mathbf{p}^2/2$ can be written[3]

$$M(\mathbf{p})=\psi_i(\mathbf{p})\int_{-\infty}^{\infty}dtW(t)\exp\left\{i\left[t(p^2/2+I)\right.\right.$$

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$$+\int_{\infty}^{t} dt' W(t') \Bigg] \Bigg\}. \tag{1}$$

Here $\phi_i(\mathbf{p})$ is the initial bound-state wave function in the momentum representation, and $W(t)$ is the interaction energy of the electron with the laser field,

$$W(t)=\frac{1}{c}\,\mathbf{pA}(t)+\frac{1}{2c^2}\,\mathbf{A}^2(t). \tag{2}$$

We take the electron charge to be $e=-1$, and use atomic units $\hbar=m=1$. The field is assumed to be circularly polarized, and to be given in the dipole approximation by the vector potential

$$\mathbf{A}(t)=(F/\omega)(\cos\omega t,\sin\omega t,0). \tag{3}$$

We consider ionization by a low-frequency ($\omega<I$) field in the tunneling regime, for which the Keldysh parameter is $\gamma=\omega\sqrt{2I}/F<1$. The mean oscillatory energy $U=F^2/2\omega^2$ is then automatically greater than the photon energy, and the strong-field parameter for the free electron is $z=F^2/2\omega^3>1$. In other words, typical energies in this problem are

$$\omega<I<U. \tag{4}$$

Finally, we assume that $F<F_a=(2I)^{3/2}$, i.e., that the laser field is weaker than the atomic field.

3. SUMMATION OVER PERIODS

The infinite integral in the expression (1) for the transition amplitude can be written as a sum of integrals over a sequence of periods separated at $\omega t_n=2\pi n$ (integer n):

$$M(\mathbf{p})=\sum_n B_n(\mathbf{p})\exp[i\eta_n(t)]. \tag{5}$$

The phases $\eta(t_n)$ are identical with the exponents in (1) at time t_n, and the contribution of one period is

$$B_n(\mathbf{p})=\psi_i(\mathbf{p})\int_0^{2\pi/\omega} dt W(t)\exp[i\eta_n(t)]. \tag{6}$$

The electron velocity, canonical momentum $\mathbf{p}$, and vector potential $\mathbf{A}$(t) are related by

$$\mathbf{v}(t)=\mathbf{p}+\frac{1}{c}\,\mathbf{A}(t), \tag{7}$$

which makes it possible to bring the phase of the integrand in (6) to the form

$$\eta_n(t)=\int_0^t dt[\varepsilon_{\mathbf{p}}(t)+I], \tag{8}$$

in which $\varepsilon_{\mathbf{p}}(t)$ is the time-dependent kinetic energy of the electron in the electromagnetic field:

$$\varepsilon_{\mathbf{p}}(t)=m\mathbf{v}^2(t)/2=\left(\mathbf{p}+\frac{1}{c}\,\mathbf{A}(t)\right)^2. \tag{9}$$

Taking note of (8), if we move the integration contour in (6) to the complex plane, we obtain (to within a multiplicative factor) the transition amplitude for the adiabatic Landau–Dykhne theory.[2]

The phases in (5) satisfy the recurrence relation

$$\eta(t_{n+1})-\eta(t_n)=2\pi\nu_n, \tag{10}$$

where for a complete phase excursion over a period we have introduced the convenient notation $\eta_n(2\pi/\omega)=2\pi\nu_n$. B_n and ν_n depend on the number of the period by way of the field strength $F=F_n(t_n+t)$.

We can easily sum over period numbers in two cases. First of all, consider the case of $F=$const assuming a laser pulse consisting of N periods. From (8),

$$\nu=\frac{1}{\omega}\left(\frac{\mathbf{p}^2}{2}+I+U\right). \tag{11}$$

Now B and ν are independent of n, and using (10), the sum in (5) reduces to a geometric progression. Dropping the common phase factor, we have[6]

$$M(\mathbf{p})=B(\mathbf{p})\frac{\sin(\pi N\nu)}{\sin(\pi\nu)}. \tag{12}$$

For large N, the ratio of sines in (12) can be replaced by $\delta(\nu-k)$ with arbitrary integer k. It is clear from (11) that we then have a relation identical with the energy conservation law for the kth harmonic in the conventional approach to Fourier expansion, and the transition amplitude (5) becomes a sum over quasienergy harmonics,

$$M(\mathbf{p})=B(\mathbf{p})\sum_k \delta(\nu-k). \tag{13}$$

With energy conservation borne in mind, the contribution of one period of $B(\mathbf{p})$ can be expressed in terms of Bessel functions. Thus, in a monochromatic field with constant envelope, the two numerical techniques—dividing up the time interval, expansion in harmonics—lead to the same result.

Note that in the time-domain approach, the transition amplitude to a state with momentum $\mathbf{p}$ that satisfies the energy conservation law is a coherent sum of contributions from all periods in the laser pulse. Clearly, the conservation of energy in a time-dependent but monochromatic field is in fact a coherence condition. If the pulse does not satisfy this condition, interfering terms of the various periods will cancel one another.

In the opposite limit, we have completely incoherent contributions from the various periods. The transition probability to a state with momentum $\mathbf{p}$, computed for the full duration of the laser pulse, reduces to a sum of incoherent contributions from all periods,

$$\left|\sum_n B_n(\mathbf{p})\exp(i\eta_n)\right|^2 \to \sum_n |B_n(\mathbf{p})|^2. \tag{14}$$

Here, the energy conservation law does not come up. In the absence of saturation, the probability is proportional to the pulse width (the number of significant terms in the sum).

Contributions from neighboring periods will be incoherent if the phase assigned to the period $2\pi\nu_n$ differs by a quantity of order unity between the two neighbors, $\delta\nu=\nu_{n+1}-\nu_n\sim 1$. Since $\delta\nu\sim U/\omega^2\tau$, the mean oscillatory

energy should rise in an optical period to something of the order of the laser photon energy. The criterion for total incoherence can also be written in the form

$$z > \omega\tau, \tag{15}$$

i.e., the strong-field parameter z must be at least of the order of the number of optical periods in the laser pulse. It is worth noting that this criterion does not depend on the electron energy or ionization potential, and it will hold under conditions that are not at all exotic, especially in the infrared. For example, a CO_2 laser with intensity 10^{14} W/cm^2 will have $z \sim 10^4$, and according to (15), the pulse need be no more than 100 ps wide. The criterion (15) and the requirement $F < F_a$ constrain the field from opposite directions, and they are compatible if $\omega\tau < (2I/\omega)^3$. For CO_2 emission and $I = 13.6$ eV, the condition holds down to pulse widths of some tenths of a nanosecond.

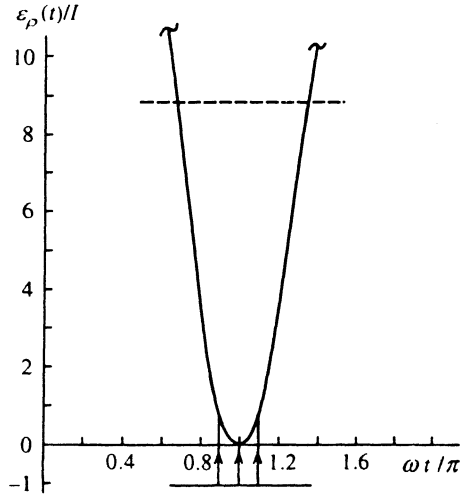

FIG. 1. Quantum transition from level I to a continuum state with $p_z = 0$, $p_\perp = p_F$, and kinetic energy (solid curve) $\varepsilon_p(t)$ for the case in which the mean oscillatory energy (dashed curve) is $U/I = 9$. Then $\gamma = 1/3$, the mean kinetic energy is $18\,I$, and the peak kinetic energy is $36\,I$. Vertical arrows mark the range in which the transition actually takes place.

4. QUANTUM TUNNELING IONIZATION TRANSITION

We now discuss the contribution of a single period to the transition amplitude. By virtue of (4), the exponential in the integrand of (6) oscillates rapidly, in general, but the oscillations are unevenly distributed over a period. The time dependence of the kinetic energy modulates the instantaneous frequency of oscillation, which equals the derivative of the phase. With $\mathbf{A}(t)/c = \mathbf{p}_F(t)$, we can write the latter in the form

$$\varepsilon_{\mathbf{p}}(t) + I = [\mathbf{p}_\perp + \mathbf{p}_F(t)]^2/(2\omega) + (I + p_z^2/2)/\omega. \tag{16}$$

The transverse component of the velocity (in the plane of polarization) comprises the constant mean momentum $\mathbf{p}_\perp$ and the field component $\mathbf{p}_F(t)$, which has invariant length $p_F = F/\omega$ and rotates uniformly at the field frequency ω. The kinetic energy reaches a minimum once per period, when the spin angular momentum is directed opposite the mean. The neighborhood of that point makes the main contribution to the transition amplitude into a state of given momentum.

If we consider the full momentum spectrum, we find that the absolute minimum of the oscillation frequency, which is equal to the ionization potential I, is attained at momenta with $p_z = 0$, arbitrary direction in the plane of polarization, and $p_\perp = p_F$, whereupon the transverse velocity can vanish. These are precisely the momenta at which the photoelectron spectrum peaks in a circular field.

Near a kinetic energy minimum, $\omega t = \pi + \omega t'$ and $\omega t' \ll 1$. Here we can expand (16), retaining the quadratic term:

$$\varepsilon_{\mathbf{p}}(t) + I = \lambda + \tfrac{1}{2} p_\perp p_F (\omega t')^2. \tag{17}$$

The minimum oscillation frequency is

$$\lambda = (p_\perp - p_F)^2/2 + I + p_z^2/2. \tag{18}$$

Substituting (17) into (6) and extending the limits of integration to $\pm\infty$, the contribution due to a single period becomes

$$B(\mathbf{p}) = 2\pi \left(\frac{\omega^2 p_F p_\perp}{2}\right)^{-1/3} \mathrm{Ai}(q)\,\psi_i(\mathbf{p})\,W(\pi)\,e^{i\pi\nu}, \tag{19}$$

where the argument of the Airy function is

$$q = \lambda/(\omega^2 p_F p_\perp/2)^{1/3}. \tag{20}$$

At $q \gg 1$, we can employ the asymptotic expansion

$$\mathrm{Ai}(q) = \frac{1}{2\sqrt{\pi}}\, q^{-1/4} \exp\left(\frac{-2q^{3/2}}{3}\right) \tag{21}$$

The properties of the photoelectron spectrum stemming from (19)–(21) are discussed in the next section.

The principal contribution to the integral (6) comes from the time interval t_0 over which the transition rate varies from a minimum value λ to a quantity of order λ. This interval can be viewed as the duration of the quantum transition. When (4) holds—all the more so, when the inequalities are sharp—the frequency of oscillations near the spectral peak varies widely over the course of a period, from values of order U to a minimum $I \ll U$. The time t_0 is therefore a small fraction of the field period, and the quantum transition is a rapid one. Assuming that $p_\perp \sim p_F$ and $\lambda \sim I$ in (17), we find $t_0 = \sqrt{2I}/F$. The duration of the quantum transition is thus identical to the tunneling time through a potential barrier, and $\omega t_0 = \gamma$ equals the standard Keldysh parameter.

These calculations give rise to the simple and transparent physical model of ionization sketched out in Fig. 1. Ionization by a strong low-frequency field in the tunneling regime ($\gamma < 1$) is fast, confined to the duration of the quantum transition from the energy level I of the initial bound state to a continuum state characterized by quantum number $\mathbf{p}$ and time-dependent energy $\varepsilon_{\mathbf{p}}(t) = m\mathbf{v}^2(t)/2$. This picture is analogous to an electronic transition between time-dependent molecular terms. The Landau–Zener quantum transition scheme also holds in a linearly polarized field. Only the time dependence of the kinetic energy changes: in contrast to Fig. 1, it has two minima per period for nonzero momentum.

Viewed from the perspective of the current study, confirmations of the classical ionization model,[7] in which the

electron is instantaneously ejected from the atom and has nonzero velocity at that instant, determine the position of the peak in the photoelectron spectrum.

Formally, the instantaneous frequency of oscillations (16) is modulated because of the cross term in the expression for $\mathbf{v}^2(t)$, which is a periodic function of time. A Fourier expansion "blurs" the effects of the cancellation of kinetic energy terms over many harmonics, and clouds the physical picture. For the cancellation to be evident, it is also important to preserve the $\mathbf{A}^2$ term.

5. THE SPECTRUM FOR SHORT PULSES

We can find the momentum distribution of photoelectrons for the tunneling regime of ionization by a strong low-frequency field in the constant-envelope case by substituting (19) into (13) and calculating the transition rate in the usual manner. The resulting expressions agree with previously derived results for this limit—results obtained in a different way. For example, the Airy function in (19) and in Ref. 8 agree if we set $p_\perp = p_F$ in the denominator of (20) and take $I = 1/2$. A tunneling spectrum proportional to the exponential in (21) was obtained in Ref. 9 for the ionization of atoms. For circular polarization, the energy spectrum peaks at $\varepsilon = \varepsilon_F$ with a width[9]

$$\Delta\varepsilon = \sqrt{\frac{2I\omega}{\gamma^3}}. \tag{22}$$

Note that this expression actually determines the spectral width if the position of the peak remains fixed.

Let us consider the energy spectrum in a strong field with a variable envelope when (15) holds, and the probability is calculated according to (14). The energy dependence of the contribution made by the nth period to (19) is the same as in a field with constant amplitude F_n. Variations in the amplitude of the field from period to period not only change the width (22), they also shift the position of the peak. After summing over all periods, the resulting distribution can become substantially wider if the displacement of the peak during the pulse duration exceeds the width of the original distribution by one period.

In a laser pulse with peak field $F(0)$, the position of the peak B ranges from 0 to $\varepsilon_F(0) = F^2(0)/2\omega^2$. It would be incorrect, however, to consider this quantity to be the spectral width. It must be borne in mind that the probability, which is proportional to $\exp[-2F_a/3F(t_n)]$, decreases rapidly as the field drops. The probability will be comparable to the peak value only as long as the field remains within $\delta F \sim F^2(0)/F_a$ of $F(0)$. This much of a variation in the field shifts the peak by approximately

$$\delta\varepsilon_F = F^3(0)/\omega^2 F_a. \tag{23}$$

If the ratio of the width (22) to the displacement (23) is less than unity,

$$\Delta\varepsilon_\perp / \delta\varepsilon_F = \sqrt{\frac{2I\gamma^3}{\omega}} < 1, \tag{24}$$

the spectral width will be determined by the displacement of the peak, and it will then be greater than in a field of constant amplitude. The ratio (24) is to be calculated at $F = F(0)$. If ionization is saturated, then the field $F(0)$ in (23) and (24) is taken to be the saturation field. For $\omega = 0.1$ eV and $I = 13.6$ eV, (24) holds when $\gamma < 0.15$.

6. CONCLUSION

The present calculation demonstrates that tunneling ionization in the Keldysh–Faisal–Reiss model can incontrovertibly be considered a Landau–Zener transition from a bound state to a continuum state with definite momentum and varying energy, the latter being the kinetic energy of a classical electron in an electromagnetic field. A time-domain approach (with no Fourier expansions) to the ionization calculation and the consequent transparent interpretation of the quantum transition can provide the basis for new modifications of the Keldysh–Faisal–Reiss model that take account of the Coulomb field influence on the final state. One task of paramount importance that was not considered in the present work is an investigation of the multiphoton regime in short pulses. From an experimental standpoint, it would be interesting to examine the spectral width in the tunneling regime more closely.

The author thanks N. B. Delone, V. P., Kraĭnov, and S. L. Chin for stimulating discussions.

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Translated by Marc Damashek