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On the ionization of atoms by electromagnetic radiation

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Abstract

The Goeppert-Mayer and Kramers-Henneberger transformations are examined for a bound charge in electromagnetic radiation in the non-relativistic approximation. It is shown that the consistent inclusion of the initial conditions ensures the equivalence of the dipole hamiltonian and the standard non-relativistic hamiltonian. These results offer the possibility of following the time evolution of the charge wavefunction with both the structural interaction (which ensures the bound state) and the interaction provided by the radiation. It is shown that after a longer or shorter time (depending on the strength of the electric field) since switching on the radiation the bound charges are set free; in these conditions the most energetic charges may tunnel through a possible potential barrier (*e.g.*, Coulomb barrier), or be rescattered, while the remaining charges are rearranged and the process is resumed. This new picture is applied to the ionization of atoms with a Coulomb potential barrier. The results differ from the well-known ionization probability obtained by "quasi-classical" tunneling through "classically unavailable" non-stationary states (imaginary-time tunneling). Extension of the approach to other applications involving radiation-induced charge emission from bound states is discussed, like ionization of molecules, atomic clusters or proton emission from atomic nuclei. Results for a static electric field are included.

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Key words: ionization by electromagnetic radiation; dissociation of bound states; fragmentation; tunneling

The investigation of the laser-matter interaction was focused since the beginning on the radiation-induced atom ionization.[1, 2] Originally, the transitions between the atomic non-stationary states have been approached by time-dependent perturbation theory. Keldysh[3] noticed that such an approach implies a "quasi-classical" tunneling through states which are not allowed by the classical dynamics.[4] Later, it was realized[5] that the tunneling formalism involves in fact a unitary transformation known as the Kramers-Henneberger transformation.[6, 7] All these calculations are of limited validity and imply necessarily simplifying approximations which affect the results. Typically, they neglect the dynamics of the electrons inside the atoms in the presence of the radiation field, which results in ionization probabilities proportional to $e^{-const/E}$, where E is the strength of the electric field[8]-[11] (this is a well-known result, valid also for static fields[4]). The need of a time-evolving picture of radiation-induced atom ionization has been often emphasized.[12, 13]

We attempt here to avoid such drawbacks. First, we show that a consistent use of the unitary transformations of the Goeppert-Mayer and Kramers-Henneberger type is needed in order to

ensure the equivalence of the dipole hamiltonian with the standard non-relativistic hamiltonian; in this equivalence the initial conditions play an essential role, a circumstance which offers the opportunity to follow the time evolution of the wavefunction with both the structural interaction (which may be responsible for the charge bound state) and the interaction caused by the radiation. Second, it is shown that, after a lapse of time, the well-known radiation-dressed atomic interaction resulting from the Kramers-Henneberger transformations may vanish, setting the electrons free to attempt to penetrate a possible potential barrier associated with the ionization. This lapse of time is short for high-intensity radiation and long for weak radiation fields. During the long time till the tunneling may appear in weak fields, the electrons oscillate between non-stationary states, leading to well-known multi-photon absorption and production of high-order electromagnetic harmonics. During the tunneling, or the rescattering, the remaining electrons are rearranged and the process is resumed. We apply this new picture to the ionization of atoms, assuming, for convenience, (non-relativistic) single-electron states in the atomic mean-field and a Coulomb potential barrier. The possibility of extension of the results to multiply-charged ions, ionization of large molecules, atomic clusters, including correlation effects, or to proton emission from atomic nuclei is discussed.

To start with, we assume a charge q with mass m in the atomic potential $V(\mathbf{r})$ in the presence of an electric field $\mathbf{E}$; in the non-relativistic approximation the dipole hamiltonion is

$$H_d = H_0 - q\mathbf{r}\mathbf{E} \ , \ \ H_0 = \frac{1}{2m}p^2 + V(\mathbf{r}) \ \ , \tag{1}$$

where $\mathbf{r}$ denotes the relative position (with respect to the center-of-mass) and $\mathbf{p}$ is the charge momentum. We assume that the electric field $\mathbf{E}$ is a radiation field; in the non-relativistic approximation we may limit ourselves to its time dependence; consequently, we assume a typical component of the electric field of the form $\mathbf{E} = \mathbf{E}_0 \sin\omega t$, where ω is the radiation frequency (linear polarization) and t denotes the time. We consider the associated Schrodinger equation $i\hbar\partial\psi/\partial t = H_d\psi$ and introduce the unitary transformation

$$\psi = e^{-\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t - 1)}\phi \ ; \tag{2}$$

we note that $\psi(t=0) = \phi(t=0)$ and the interaction $-q\mathbf{r}\mathbf{E}_0 \sin\omega t$ vanishes at $t = 0$. The transformation given by equation (2) leads to

$$\mathbf{p} \to \widetilde{\mathbf{p}} = e^{\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t - 1)}\mathbf{p}e^{-\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t - 1)} = \mathbf{p} - \frac{q}{\omega}\mathbf{E}_0(\cos\omega t - 1) \ ; \tag{3}$$

since we may view the electric field $\mathbf{E}$ as derived from the vector potential $\mathbf{A} = \mathbf{A}_0 \cos\omega t$, $\mathbf{A}_0 = c\mathbf{E}_0/\omega$, the above equation becomes

$$\widetilde{\mathbf{p}} = \mathbf{p} - \frac{q}{c}\mathbf{A}_0(\cos\omega t - 1) = \mathbf{p} - \frac{q}{c}(\mathbf{A} - \mathbf{A}_0) \tag{4}$$

and the transformed hamiltonian reads

$$\widetilde{H}_d = e^{\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t - 1)}H_d e^{-\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t - 1)} = \frac{1}{2m}\left[\mathbf{p} - \frac{q}{c}(\mathbf{A} - \mathbf{A}_0)\right]^2 + V(\mathbf{r}) \ \ , \tag{5}$$

with the associated Schrodinger equation $i\hbar\partial\phi/\partial t = \widetilde{H}_d\phi$; we recognize in equation (5) the standard non-relativistic hamiltonian with $\mathbf{A} - \mathbf{A}_0$ instead of the usual $\mathbf{A}$. The presence of the constant $\mathbf{A}_0$ does no affect the classical equations of motion, and, of course, it does not affect the equations of the electromagnetic radiation (it is a gauge transformation); however, we shall see shortly that it has important effects on the quantum-mechanical dynamics of the charge. The transformation given by equation (2) is known as the Goeppert-Mayer transformation.[14]

Let us write

$$\widetilde{H}_d = H_0 - \frac{q}{mc}(\mathbf{A}-\mathbf{A}_0)\mathbf{p} + \frac{q^2}{2mc^2}(\mathbf{A}-\mathbf{A}_0)^2 \tag{6}$$

and continue with the unitary transformation

$$\phi = e^{-\frac{iq^2}{8mc^2\hbar\omega}A_0^2(\sin 2\omega t - 8\sin\omega t + 6\omega t)} e^{\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}\chi \ ; \tag{7}$$

it leads to the Schrodinger equation

$$i\hbar\frac{\partial\chi}{\partial t} = \left[\frac{1}{2m}p^2 + \widetilde{V}(\mathbf{r})\right]\chi \tag{8}$$

and the radiation-dressed atomic potential

$$\widetilde{V}(\mathbf{r}) = e^{-\frac{q}{m\omega^2}(\sin\omega t-\omega t)\cdot\mathbf{E}_0 grad} V(\mathbf{r}) \ . \tag{9}$$

Therefore, collecting all these transformations, the original wavefunction can be written as

$$\psi = e^{-\frac{iq^2}{8m\hbar\omega^3}E_0^2(\sin 2\omega t - 8\sin\omega t + 6\omega t)} e^{-\frac{i}{\hbar\omega}q\mathbf{r}\mathbf{E}_0(\cos\omega t-1)} e^{\frac{iq}{m\hbar\omega^2}(\sin\omega t-\omega t)\cdot\mathbf{E}_0\mathbf{p}}\chi \ ; \tag{10}$$

we note that $\psi(t=0) = \chi(t=0)$ and the electromagnetic interaction vanishes at $t=0$ both in the original dipole hamiltonian H_d ($-q\mathbf{r}\mathbf{E}_0\sin\omega t \mid_{t=0} = 0$) and the standard non-relativistic hamiltonian given by equation (6) ($(\mathbf{A}-\mathbf{A}_0)\mid_{t=0} = \mathbf{A}_0(\cos\omega t - 1)\mid_{t=0} = 0$); this establishes the equivalence of the two hamiltonians.

The above unitary transformations are the well-known Kramers-Henneberger transformations, including the radiation-dressed potential $\widetilde{V}(\mathbf{r})$;[5]-[7] the difference with respect to their usual form consists in the introduction of the initial conditions, reflected in the presence of ωt in the exponent $\sin\omega t - \omega t$ in equation (10). Usually, this contribution is omitted; however, it is this factor which ensures the equivalence of the transformed hamiltonians.

We proceed now to apply these results to the ionization of atoms; we assume that in the absence of the radiation field the potential $V(\mathbf{r})$ is the mean-field potential which generates atomic bound single-electron states. Let us assume that the electric field $\mathbf{E}$ is directed along the z-axis; then, equation (9) gives

$$\begin{aligned}\widetilde{V}(x,y,z) = V - \tfrac{qE_0}{m\omega^2}(\sin\omega t-\omega t)V_1 + \tfrac{q^2E_0^2}{2m^2\omega^4}(\sin\omega t-\omega t)^2V_2 + ... = \\ = V\left(x,y,z-\tfrac{qE_0}{m\omega^2}(\sin\omega t-\omega t)\right) \ ,\end{aligned} \tag{11}$$

where $V_1 = \partial V/\partial z$, $V_2 = \partial^2 V/\partial z^2$, ... We can see that the potential $\widetilde{V}$ at the position of the charge is the original potential V at coordinate $z - (qE_0/m\omega^2)(\sin\omega t - \omega t)$; it follows that for a sufficiently long time this potential may vanish (due to the ωt-term). We introduce the parameter $\xi = qE_0/m\omega^2 a$, where a is of the order of the dimension of the atom. We can see that for $\xi \ll 1$ the charge q (the electron) suffers for a long time the small oscillating effects of the radiation while moving in the atomic potential; in these circumstances we may have multi-photon absorption and emission of high-order harmonics, from terms of the form $\sin^n\omega t$ in $\widetilde{V}$; this is a well-known effect;[15]-[18] we note that all these occur for weak fields ($\xi \ll 1$). It is convenient to write the parameter ξ as $\xi = \eta(\lambda/a)$, where $\eta = qA_0/mc^2$ and λ is the radiation wavelength; in order to preserve the non-relativistic approximation we should have $\eta \ll 1$. On the other hand, for $\xi \gg 1$, after a certain, short, lapse of time τ, the potential $\widetilde{V}$ at the position of the charge is vanishing, and the charge is set free. The conditions $\xi > 1$ and $\eta < 1$ are satisfied for the radiation

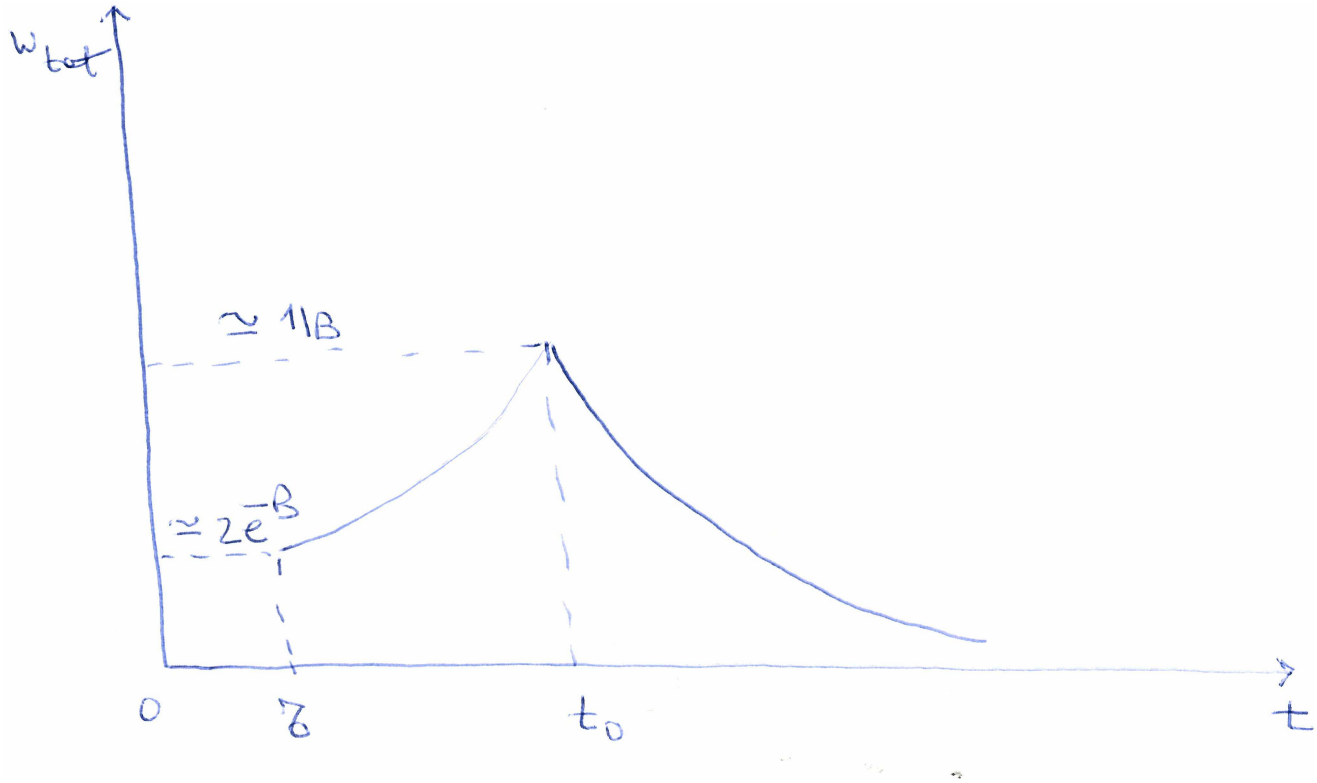

Figure 1: Schematic representation of the total tunneling probability w_{tot} (in arbitray units) *vs* time t for $A \ll 1 \ll B$ (the notations are defined in text).

intensity I in the range $10^{12} w/cm^2 < I < 10^{18} w/cm^2$ for electrons in atoms ($a = 10^{-8} cm$) and optical radiation ($\omega = 10^{15} s^{-1}$, $\lambda \simeq 10^4 cm$). We can see that for moderate laser intensities the parameter ξ is large ($\xi \simeq 10^3$ for $I = 10^{18} w/cm^2$). Under these circumstances the time τ is given by

$$\xi(\omega t - \sin \omega\tau) \simeq \frac{1}{6}\xi(\omega\tau)^3 = 1 \ . \tag{12}$$

To continue we use a simplified atomic model. In the lapse of time τ the electrons are set free; they have momenta $\mathbf{p}_n$ and kinetic energies $\varepsilon_n = p_n^2/2m$, where n is a generic notation for the electron states; we may leave aside the orbital motion and denote by $\mathbf{p}_{rn}$ the radial momentum and by ε_{rn} the radial energy. Let $\mathbf{p}_r$ and $\varepsilon_r = p_r^2/2m$ be the highest radial momentum and, respectively, the highest radial energy; they correspond to the total momentum $\mathbf{p}$ and, respectively, total energy $\varepsilon = p^2/2m$ (a degeneration may exist, which can be included). This electron may tunnel through a possible (centrally-symmetric) potential barrier $U(r)$ from r_0 to r_1, where $U(r_1) = \varepsilon_r$.[1] The relevant factors in the wavefunction ψ given by equation (10) are

$$e^{-\frac{iq}{m\hbar\omega^2}(\omega t - \sin\omega t)E_0 \cos\theta \cdot (p_1 - p_0) + \frac{i}{\hbar}\int_{r_0}^{r_1} dr \cdot p_r(r)} \ , \tag{13}$$

where $p_r(r) = \sqrt{2m\left[\varepsilon_r - U(r)\right]}$, $p_{0,1} = p(r_{0,1}) = \sqrt{2m\left[\varepsilon - U(r_{0,1})\right]}$ and θ is the angle between $\mathbf{E}_0$ and $\mathbf{p}$. It is easy to see that p_1 is always real; it follows that the tunneling probability (transmission coefficient) is given by $w = e^{-\gamma}$, where

$$\begin{aligned} &\gamma = A\xi(\omega t - \sin\omega t)\cos\theta + B \ , \\ &A = \tfrac{2a|p_0|}{\hbar} \ , \ \ B = \tfrac{2}{\hbar}\int_{r_0}^{r_1} dr\, |p_r(r)| \end{aligned} \tag{14}$$

[1] This is not so for atoms, where the potential is reduced by radiation; the tunneling holds for charge ejection from atomic nuclei.

and $|p_0| = \sqrt{2m\,[U(r_0) - \varepsilon]}$ and $|p_r(r)| = \sqrt{2m\,[U(r) - \varepsilon_r]}$; this equation is valid for $t > \tau$.

The estimation of the coefficients A and B depends on the atomic model and the potential barrier $U(r)$. Let us assume a Coulomb barrier $U(r) = Zq^2/r$, where, for the sake of the generality, we consider an ion with charge $-Zq$. The tunneling conditions $r_0 < r_1$ and $|p_0|$ real and positive lead to the inequalities

$$\frac{\varepsilon_r \varepsilon r_0}{Zq^2} < \varepsilon_r < \frac{Zq^2}{r_0} \quad , \tag{15}$$

which imply $\varepsilon_r \varepsilon < (Zq^2/r_0)^2$; this inequality shows that the tunneling occurs for peripheral electrons ("valence" electrons), whose kinetic energy is sufficiently small, as in a hydrogen-like model of atoms (or for Rydberg states); this is an expected result. In order to illustrate the analytical calculations we use now a rather crude approximation which consists in neglecting the kinetic energies in $|p_0|$ and $|p_r(r)|$ and setting $r = a$; this approximation (suggested by the inequalities (15)) does not affect the qualitative conclusions described below; doing so, we get the approximate expressions

$$A \simeq 2\sqrt{2Za/a_H} \ , \ \ B \simeq 4Z\sqrt{2q^2/a_H \varepsilon_r} \gg 1 \tag{16}$$

for the coefficients A and B, where $a_H = \hbar^2/mq^2$ is the Bohr radius; we can see that, within this approximation, the tunneling probability is dominated by the coefficient B (which does not include the radiation field); if we use the localization energy $\varepsilon_r = \hbar^2/ma^2$ for the kinetic energy ε_r the coefficient B becomes $B = 4\sqrt{2}Za/a_H$.

The motion proceeds along the path of the minimal phase variation, such that, in equation (14) we have the condition $\gamma > 0$, *i.e.*,

$$\cos\theta > \cos\theta_0 = -\frac{B}{A\xi(\omega t - \sin\omega t)} \ ; \tag{17}$$

we can see that for $\tau < t < t_0 = (B/A)^{1/3}\tau$ the ejection of the charge proceeds at all angles (since $B > A$), while for $t > t_0 = (B/A)^{1/3}\tau$ it proceeds only at angles $0 < \theta < \theta_0$; for large times $\theta_0 \to \pi/2$ and the emission occurs mainly along the direction of the electric field, as expected. We can define a total probability w_{tot} by integrating over angle θ; we get

$$w_{tot} = \frac{2\sinh\left[A\xi(\omega t - \sin\omega t)\right]}{A\xi(\omega t - \sin\omega t)} e^{-B} \ , \ \ \tau < t < t_0 \tag{18}$$

and

$$w_{tot} = \frac{1 - e^{-A\xi(\omega t - \sin\omega t) - B}}{A\xi(\omega t - \sin\omega t)} \ , \ \ t > t_0 \ ; \tag{19}$$

we can see that the probability is dominated by the fast process of tunneling occurring in the short time interval $\tau < t < t_0$ (equation (18)); at large times the probabilitiy is vanishing.

The number of ejected electrons per unit area in the time interval dt is $w_{tot} v_r dt$, where v_r $(= p_r/m)$ is their radial velocity (the wavefunction is normalized to the unit volume); therefore, the total flux of ejected electrons is obtained by integrating $w_{tot} v_r$ from $t = \tau$ to infinity; it is easy to see that the integral $\int_\tau^\infty dt w_{tot}$ may be approximated by $\simeq t_0/B$ for large values of the coefficient B. The total probability w_{tot} given by equations (18) and (19) is represented schematically in Fig. 1. We can see that the radiation effect is included in the time t_0 (time τ); for high radiation fields (increasing ξ) the time t_0 (time τ) and the emission flux decrease; for high-intensity radiation the emission of high-order harmonics may be controlled by the very short time t_0.

After the time τ one or more electrons may tunnel (together or sequentially), leading to multiply-charged ions; indeed, the Kramers-Henninger transformations can be applied straghtforwardly to several interacting charges in radiation field (the corresponding wavefunction must be properly symmetrized). However, when the tunneling process starts the electronic core suffers a reconfiguration (rearrangement) process and the potential $V(\mathbf{r})$ is modified; this is the well-known process of core "shake-up" (or core excitation).[17]-[19] As a consequence of this reconfiguration process, the condition of setting the electrons free given by equation (11) is not valid anymore; a new bound state is formed and a new transformation process begins for the modified potential $V(\mathbf{r})$.

The tunneling probability w given above is a transmission coefficient (we can check that $w < 1$); with probability $1 - w$ the electron is reflected from the potential barrier; in these conditions the electronic core is "shaken-up" and, if the radiation field is weak, the electron resumes its process of multi-photon absorption and emission of high-order harmonics, untill it may be rescattered back to the atomic core; these are the well-known recollision processes.[15]-[18]

We note that the above calculations are for linearly polarized radiation; it is easy to see that similar calculations can be done for circular (or elliptical) polarization.

The probability w_{tot} is a function of $\xi(\omega t - \sin\omega t)$; for higher-intensity radiation (increasing ξ) the emission of the charge is faster; in the time interval $\tau < t < t_0 = (B/A)^{1/3}\tau$ the probability increases (equation (18)), but the total flux decreases, as a consequence of decreasing t_0. An interesting question arises here, related to very high-intensity radiation in the so-called relativistic regime, where $\eta \gg 1$. As long as the bound state of the charge subsists, the motion is, practically, non-relativistic; this means that the electromagnetic momentum $\mathbf{p}$ is sufficiently large to reduce to a large extent the contribution $q\mathbf{A}/c$, such that the velocity is small; the above non-relativistic formalism may be applied. However, this situation lasts a very short time (since $\xi \gg 1$); the barrier may be penetrated very rapidly (practically the barrier may be neglected for extremely high-intensity radiation), and the charge is injected in the high-intensity radiation, where it is rapidly accelerated up to relativistic velocities.[21]-[23]

Finally, we include here similar calculations for a static uniform electric field $\mathbf{E}$, which may be viewed as being derived from a vector potential $\mathbf{A} = -c\mathbf{E}t$. The wavefunction is

$$\psi = e^{-\frac{iq^2E^2t^3}{6\hbar m}} e^{\frac{iqt}{\hbar}\mathbf{Er}} e^{-\frac{iqt^2}{2\hbar m}\mathbf{Ep}} \chi \quad , \tag{20}$$

where χ satisfies the Schrodinger equation

$$i\hbar\frac{\partial\chi}{\partial t} = \frac{1}{2m}p^2\chi + \widetilde{V}(\mathbf{r})\chi \tag{21}$$

with the transformed potential given by

$$\widetilde{V}(\mathbf{r}) = e^{\frac{qt^2}{2m}\mathbf{E}grad} V(\mathbf{r}) \ . \tag{22}$$

It is easy to see that the dissociation occurs in the time interval

$$\tau = \sqrt{\frac{2ma}{qE}} \tag{23}$$

and the tunneling probability is given by $e^{-\gamma}$, where

$$\gamma = A(t/\tau)^2\cos\theta + B \tag{24}$$

and

$$\cos\theta > \cos\theta_0 = -\frac{B}{A(t/\tau)^2} \ . \tag{25}$$

For $\tau < t < t_0 = (B/A)^{1/2}\tau$ the emission proceeds in all directions, while for $t > t_0$ it occurs mainly in the forward direction. The total probability is

$$w_{tot} = \frac{2\sinh\left[A(t/\tau)^2\right]}{A(t/\tau)^2} e^{-B} \ , \ \tau < t < t_0 \tag{26}$$

and

$$w_{tot} = \frac{1 - e^{-A(t/\tau)^2 - B}}{A(t/\tau)^2} \ , \ t > t_0 \ . \tag{27}$$

We can see that these analytical expressions for the tuneling probability differ from the usual expression of the from $e^{-const/E}$. The "relativistic" parameter η may be cast, in this case, in the form $\eta = \sqrt{2qEa/mc^2}(t/\tau)$.

In conclusion, we may say that by means of a suitable unitary transformation of the Goeppert-Mayer type it is shown that the dipole hamiltonian and the standard non-relativistic hamiltonian of charges in electromagnetic radiation are equivalent. The initial condition plays an important role in this equivalence, which implies, in unitary transformations of the Kramers-Henneberger type, a gauge-like contribution to the phase of the wavefunction which may have physical consequences; although this effect may be related to the well-known Aharonov-Bohm effect, it seems more likely that both effects originate in the non-locality of the wavefunction. Using unitary transformations of the Kramers-Henneberger-type it is shown that the radiation-dressed structural interaction (which may be responsible of the charges bound state) favours the dissociation of the bound state, which may lead to ionization, possibly by tunneling. The atom ionization probability is given and explicitly estimated in a simplified atomic model with a Coulomb potential barrier. The new picture of atomic ionization described in this paper implies an explicit time evolution of the wavefunction with both structural and radiation interaction, which exhibits different behaviour at different stages: in time τ after turning-on the radiation the bound charges are set, practically, free; in the next time interval till t_0 the tunneling may occur, followed by an ionization probability which becomes extinct for long times. The times τ and t_0 are explicitly estimated.

The approach presented here can be extended to the ionization of molecules[24, 25] or atomic clusters,[26] or to proton emission from atomic nuclei,[27] or even ion emission from molecules. For electrons in atoms the conditions of ionization and non-relativsitic approximation imply a radiation intensity in the range $10^{12}w/cm^2 < I < 10^{18}w/cm^2$, for atomic dimensions of the order $a = 10^{-8}cm$ and optical radiation with frequency $\omega = 10^{15}s^{-1}$; similar conditions lead to $10^{8}w/cm^2 < I < 10^{24}w/cm^2$ for proton emission from atomic nuclei and $10^{18}w/cm^2 < I/A^2 < 10^{24}w/cm^2$ for ion emission from molecules, where A is the mass number of the ion.

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