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A note on the electromagnetic interaction. Radiation-induced fragmentation (dissociation) of quantum-mechanical assemblies

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Abstract

It is shown that some difficulties arising in the non-relativistic quantum-mechanical treatment of a charge in the field of the electromagnetic radiation can be solved by analyzing the quantization of the motion starting with the classical mechanical action and the hamiltonian formalism. By means of unitary transformations of the Kramers-Henneberger type the equivalence of the "dipole" hamiltonian and the "standard" non-relativistic hamiltonian is shown and an effective (dressed) potential is derived, which includes the radiation effects on the original structural (bound-state) potential. It is shown that for low-intensity radiation there appear multi-photon absorption and high-order harmonics generation, while moderate- or high-intensity radiation favours the fragmentation of the bound states of charges (dissociation). The fragmentation occurs through the complete removal, after a lapse of time, of both the radiation and the structural interaction; the process continues, partly, with the fragments penetrating a possible potential barriers and, partly, with the recombination of the fragments (recollisions). The fragmentation (dissociation) rate is computed explicitly for one charge penetrating through Coulomb barriers. Applications are discussed to ionization of atoms and large molecules, radiation-induced proton emission from atomic nuclei, ion emission from molecules or fragmentation of atomic clusters.

Introduction. The interaction of laser radiation with matter was focused since the beginning on the radiation-induced atom ionization. Thereafter, the problem was extended to the ionization of the molecules or atomic clusters in laser radiation, radiation-induced proton emission from atomic nuclei or even ion emission from molecular aggregates.[1]-[4] In general, we can formulate the problem of the ejection of a charge, or several charges, from a bound state, as caused by electromagnetic radiation. This problem can be called radiation-induced fragmentation (dissociation) of bound states. Originally, the problem was approached by time-dependent perturbation theory, taking into account the fact that there not exist stationary states. Keldysh[5] noticed that such a calculation implies a "quasi-classical" tunneling[1] through a potential barrier that separates the bound state from the free state.[6] Later, it was realized that the formal tunneling approach involves in fact a unitary transformation known as the Kramers-Henninger transformation.[7]-[9][2]

[1]The "quasi-classical" tunneling[6] implies an exponent $-(2/\hbar)Im\int(-Edt+\mathbf{p}d\mathbf{r})$, where $dS=-Edt+\mathbf{p}d\mathbf{r}$ is the classical action (E being the energy and $\mathbf{p}$ being the momentum), the integral being performed near the point of "classically impossible motion"; the motion should proceed with a large increase in the mechanical action, which means an adiabatic interaction, lasting a long time, and a long path with large momentum, leading to a small exponent. As we shall see in this paper, these conditions are not met especially for high-intensity radiation.

[2]In Ref. [8] the transformed structural (bound-state) potential is applied to a harmonic oscillator which interacts with the radiation.

All these calculations imply simplifying approximations which affect the results. Typically, the results have an "exponential accuracy", *i.e.* they include only small exponentials and account at most only for the potential barrier generated by radiation.[10]-[13] In addition, they neglect details of the charge dynamics. We show here that a consistent inclusion of the initial conditions in the Kramers-Henninger transformation may remove completely both the radiation and the structural (bound-state) interaction, leading to the fragmentation (dissociation) of the bound state; the process continues, partly, with the (standard) tunneling through a possible potential barrier generated both by the radiation and the structural interaction and, partly, with the recombination of the fragments (recollisions). We apply this procedure to the ejection of a charge from a bound state through a Coulomb barrier and discuss various other cases of fragmentation, like proton emission from atomic nuclei or ion emission from molecules.

A few other technical points related to charge motion in electromagnetic radiation are clarified. First, we should be aware that, even if the radiation has a "relativistic" intensity, originally, as long as the bound state persists (for a very short time), the motion of the charge is practically non-relativistic; this is achieved by increasing correspondingly the electromagnetic momentum $\mathbf{p}$ in the well-known velocity expression $\mathbf{v} = (\mathbf{p} - q\mathbf{A}/c)/m$, where q is the charge, $\mathbf{A}$ is the vector potential, m is the mass of the charge and c is the speed of light in vacuum, such that $v/c \ll 1$. The well-known intensity parameter $\eta = qA/mc^2$, which separates the non-relativistic regime ($\eta < 1$) from the relativistic regime ($\eta > 1$) is relevant for a free charge. This is why we give special attention to the non-relativistic approximation and start our considerations with this approximation. In particular, it is emphasized that the spatial dependence of the radiation field brings higher-order relativistic corrections and should be left aside within the non-relativistic aprpoximation.

Second, it is well-known that there exists two non-relativistic hamiltonians of a charge in radiation field, one which includes the electric field (the so-called "dipole" approximation) and another, which includes the vector potential (the so-called "standard" non-relativistic hamiltonian). These two hamiltonians have completely different forms. We show here, by using a unitary transformation of the Kramers-Henninger type, that the two forms are equivalent (as they should); however, the equivalence is realized only by a consistent treatment of the initial conditions. We emphasize this special requirement of the theory, because, as natural as it may appear, its infringement may lead to appreciable inconsistencies. It is this condition which ensures the removal, after some duration, of the interaction (both the electromagnetic and the structural interaction).

Hamiltonians. The mechanical action of a charge q with mass m interacting with an electromagnetic radiation field with the vector potential $\mathbf{A}$ ($div\mathbf{A} = 0$) is

$$S = \int_a^b (-mcds - \frac{q}{c}A_i dx^i) \quad , \tag{1}$$

where s is the world-line length, x^i are the coordinates, a and b are two world points and c is the speed of light in vacuum. Introducing $ds = c\sqrt{1 - v^2/c^2}dt$, where $\mathbf{v}$ is the velocity of the charge, we get

$$S = \int_a^b (-mc^2\sqrt{1 - v^2/c^2}dt + \frac{q}{c}\mathbf{A}d\mathbf{r}) \quad , \tag{2}$$

or

$$S = \int_a^b (-mc^2\sqrt{1 - v^2/c^2} + \frac{q}{c}\mathbf{vA})dt \quad , \tag{3}$$

which highlights the well-known lagrangian

$$L = -mc^2\sqrt{1 - v^2/c^2} + \frac{q}{c}\mathbf{vA} \tag{4}$$

of a charge in radiation field.

From (1) we can write also

$$S=\int_a^b(-mc^2\sqrt{1-v^2/c^2}dt-\tfrac{q}{c}\mathbf{r}d\mathbf{A})=$$

$$=\int_a^b(-mc^2\sqrt{1-v^2/c^2}dt-\tfrac{q}{c}\mathbf{r}\tfrac{\partial\mathbf{A}}{\partial t}dt-\tfrac{q}{c}\mathbf{r}\partial_\alpha\mathbf{A}dx^\alpha)= \qquad (5)$$

$$=\int_{t_a}^{t_b}(-mc^2\sqrt{1-v^2/c^2}-\tfrac{q}{c}\mathbf{r}\tfrac{\partial\mathbf{A}}{\partial t}-\tfrac{q}{c}\mathbf{r}(\mathbf{v}grad)\mathbf{A})dt\ \ ,$$

or, since $grad(\mathbf{vA})=(\mathbf{v}grad)\mathbf{A}+\mathbf{v}\times curl\mathbf{A}$ (the derivatives being taken at constant $\mathbf{v}$),[3]

$$S=\int_{t_a}^{t_b}[-mc^2\sqrt{1-v^2/c^2}+q\mathbf{r}\mathbf{E}+\frac{q}{c}\mathbf{r}(\mathbf{v}\times\mathbf{H})-\frac{q}{c}\mathbf{r}grad(\mathbf{vA})]dt\ \ , \qquad (6)$$

where $\alpha=1,\,2,\,3$. We can see the occurence of the Lorentz force in the lagrangian.

The most convenient way of getting the hamiltonian is to use equation (4), which gives

$$\mathbf{p}=\frac{\partial L}{\partial\mathbf{v}}=\frac{m\mathbf{v}}{\sqrt{1-v^2/c^2}}+\frac{q}{c}\mathbf{A}\ ; \qquad (7)$$

from $H=\mathbf{vp}-L$ we get the hamiltonian

$$H=c\sqrt{m^2c^2+(\mathbf{p}-\frac{q}{c}\mathbf{A})^2}\simeq mc^2+\frac{1}{2m}(\mathbf{p}-\frac{q}{c}\mathbf{A})^2+...\ \ , \qquad (8)$$

where the non-relativistic limit is given (it is worth noting that, formally, in the derivation of the non-relativistic hamiltonian we kept corrections up to the v^2/c^2-order). The quantization of the hamiltonioan leads to Dirac or Schrodinger equation (or Klein-Gordon equation). For instance, we get the Schrodinger equation

$$i\hbar\frac{\partial\psi}{\partial t}=\frac{1}{2m}(-i\hbar\frac{\partial}{\partial\mathbf{r}}-\frac{q}{c}\mathbf{A})^2\psi \qquad (9)$$

for the wavefunction ψ, where $\hbar$ is Planck's constant and the term mc^2 has been dropped (it gives a phase factor $e^{-\frac{i}{\hbar}mc^2t}$).

The separate dependence of the non-relativistic hamiltonian on the position $\mathbf{r}$ (included in $\mathbf{p}$) and the world coordinate $\omega t-\mathbf{kr}$ included in $\mathbf{A}$ makes difficult the solution of the Schrodinger (or Dirac) equation (ω is the radiation frequency and $\mathbf{k}$ is the radiation wavevector). However, turning back to the action function, we can get some simplifications.

The potential vector includes terms of the form $\mathbf{A}=\mathbf{A}_0\cos(\omega t-\mathbf{kr})$. The infinitesimal phase of the radiation is

$$\omega dt-\mathbf{k}d\mathbf{r}=\omega dt\left(1-\frac{\mathbf{vk}}{ck}\right)\ ; \qquad (10)$$

the charge moves in the direction of the lowest-varying phase, which means $\mathbf{vk}=vk$ ($\mathbf{v}$ and $\mathbf{k}$ parallel); the charge becomes rapidly relativistic, *i.e.* $v\simeq c$; the potential vector becomes a constant, the term $\mathbf{vA}$ is ineffective in the action, and the charge is practically a free particle (classical, or quasi-classical). In a (traveling) radiation wave the charge becomes rapidly relativistic and behaves like a free particle; it sees not anymore the field, while the reaction force is still small, up to extremely relativistic velocities where it may dominate; however, such extreme relativistic velocities are not reached for realistic conditions; when the reaction force dominates the validity

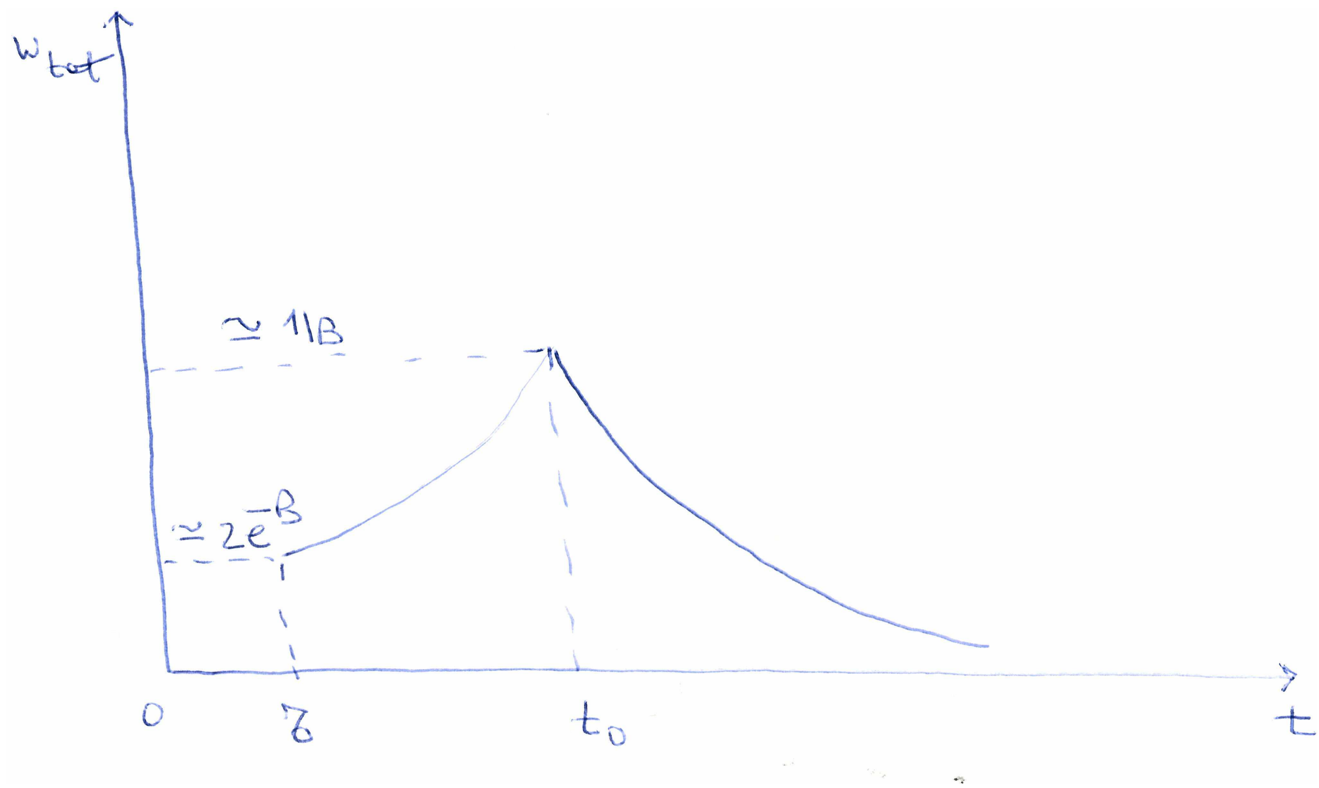

Figure 1: Schematic representation of the total tunneling probability w_{tot} (in arbitray units) *vs* time t for $A \ll 1 \ll B$ (the notations are defined in text).

of the classical electromagnetism ceases. Similar arguments can be used for an electromagnetic standing wave, which is a sum of two waves traveling in opposite directions; it is easy to see that the motion in a standing wave is possible only with small, non-relativistic, velocities.

In the non-relativistic limit the inequality $\mathbf{vk}/ck \ll 1$ is valid and we may neglect the spatial phase in the vector potential, which depends now only on ωt. This is equivalent with recognizing that the magnetic field has not any appreciable effect, and it may be neglected. From the above equations the lagrangian is

$$L = \frac{1}{2}mv^2 + q\mathbf{rE} \tag{11}$$

and the hamiltonian is

$$H_d = \frac{1}{2m}p^2 - q\mathbf{rE} \ ; \tag{12}$$

the corresponding Schrodinger equation is

$$i\hbar\frac{\partial\psi}{\partial t} = \frac{p^2}{2m}\psi - q\mathbf{rE}\psi \quad , \tag{13}$$

where $\mathbf{p} = -i\hbar\frac{\partial}{\partial\mathbf{r}}$ (the relativistic term mc^2 is left aside); in these equations the electric field has components of the form $\mathbf{E} = (\omega\mathbf{A}_0/c)\sin\omega t$. The hamiltonian H_d may be called the "dipole" hamiltonian, indicated by the suffix d. On the other hand, from equation (8) we get the non-relativistic hamiltonian

$$H_p = \frac{1}{2m}(\mathbf{p} - \frac{q}{c}\mathbf{A})^2 = \frac{p^2}{2m} - \frac{q}{mc}\mathbf{pA} + \frac{q^2}{2mc^2}A^2 \quad , \tag{14}$$

[3] In general, $grad(\mathbf{ab}) = (\mathbf{a}grad)\mathbf{b} + (\mathbf{b}grad)\mathbf{a} + \mathbf{a}\times curl\mathbf{b} + \mathbf{b}\times curl\mathbf{a}$.

which leads to the Schrodinger equation

$$i\hbar\frac{\partial\psi}{\partial t}=\frac{1}{2m}(\mathbf{p}-\frac{q}{c}\mathbf{A})^2\psi=$$
$$=\frac{p^2}{2m}\psi-\frac{q}{2mc}(\mathbf{pA}+\mathbf{Ap})\psi+\frac{q^2}{2mc^2}A^2\psi\ ; \tag{15}$$

the suffix p comes from "momentum" and the hamiltonian H_p may be called the "standard" non-relativistic hamiltonian.

Although the hamiltonians H_d and H_p given by equations (12) and, respectively, (14) look different, they are in fact equivalent; H_d is obtained by neglecting v/c in the phase of the radiation, while H_p is obtained up to corrections of order v^2/c^2 in magnitude. It is important to note that the spatial phase must be neglected in H_p, in order to keep the estimation within the same order of magnitude. Consequently, we may say that the non-relativistic limit implies only the temporal phase. Within these conditions it is easy to check that the classical equations of motion derived from both H_d and H_p have the same solutions. It is worth noting that the A^2-term in equation (14) does not contribute to the classical equations of motion, as it depends only on the time. Similar considerations are valid for static fields too. We note that the existence of a magnetic field implies relativistic effects.

Schrodinger equation. Equivalence of the hamiltonians. We pass now to the analysis of the quantum-mechanical hamiltonians $H_{d,p}$. Let us add a potential $V(\mathbf{r})$ and denote

$$H_0=\frac{1}{2m}p^2+V(\mathbf{r})\ ; \tag{16}$$

the Schrodinger equation (13) becomes

$$i\hbar\frac{\partial\psi}{\partial t}=H_0\psi-q\mathbf{r}\mathbf{E}_0\sin\omega t\cdot\psi\ \ , \tag{17}$$

where $\mathbf{E}_0=\omega\mathbf{A}_0/c$; using the unitary transform

$$\psi=e^{-\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}\phi\ \ , \tag{18}$$

we get

$$i\hbar\frac{\partial\phi}{\partial t}=e^{\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}H_0e^{-\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}\ ; \tag{19}$$

making use of

$$e^OBe^{-O}=B+[O,B]+\frac{1}{2!}[O,[O,B]]+... \tag{20}$$

for any operators O and B, we get

$$e^{\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}\mathbf{p}e^{-\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}=\mathbf{p}-\frac{q}{\omega}\mathbf{E}_0(\cos\omega t-1)=\mathbf{p}-\frac{q}{c}(\mathbf{A}-\mathbf{A}_0) \tag{21}$$

and

$$i\hbar\frac{\partial\phi}{\partial t}=e^{\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0\cos\omega t}H_0e^{-\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0\cos\omega t}\phi=\frac{1}{2m}\left[\mathbf{p}-\frac{q}{c}(\mathbf{A}-\mathbf{A}_0)\right]^2\phi+V(\mathbf{r})\phi=H_p^{'}\phi\ ; \tag{22}$$

this shows explicitly the equivalence of the hamiltonians H_d and $H_p^{'}$. It is worth noting that it is the equivalence of H_d with $H_p^{'}$ not with H_p given by equation (14); the former hamiltonian includes $\mathbf{A}-\mathbf{A}_0$, while the latter includes only $\mathbf{A}$. In writing the interaction with the radiation in the mechanical action it is immaterial a constant $\mathbf{A}_0$ in describing the radiation field, while this constant is important in the charge dynamics, through the coupling $\mathbf{pA}_0$. The transformation given by equation (18) is known as the Goeppert-Mayer transformation.[14]

It is of utmost importance to realize the relevance of including correctly the initial conditions in the unitary transformation given by equation (18). First, we note that it is our desire to remove the interaction $-q\mathbf{r}\mathbf{E}_0\sin\omega t$ to an extent as large as possible; this is why we require $\psi(t=0)=\phi(t=0)$, where the interaction $-q\mathbf{r}\mathbf{E}_0\sin\omega t\mid_{t=0}=0$ disappears for $t=0$. Second, we note that the interaction disappears in $H_p^{'}$ at $t=0$ only if $\mathbf{A}_0$ is included; we would not have the equivalence of the hamiltonians H_d and $H_p^{'}$ without the correct inclusion of the initial condition.

Similarly, it would not be convenient to take $\mathbf{A}=\mathbf{A}_0\sin\omega t$ and $\mathbf{E}=\mathbf{E}_0\cos\omega t$ ($\mathbf{E}_0=-\omega\mathbf{A}_0/c$), since then the interaction is not vansihing at $t=0$, but at $t_0=\pi/2\omega$, which would introduce a small inconvenience in calculations. We emphasize that the interaction is $-q\mathbf{r}\mathbf{E}$ and not $-q\mathbf{v}\mathbf{A}/c$, since $\mathbf{E}$ is uniquely definible (measurable), while $\mathbf{A}$ is not.

We note also that the constant term $\mathbf{A}_0$ has no effect on the classical dynamics; also, it has no effect on the quantum-mechanical dynamics as long as its contribution to the phase of the wavefunction preserves a real phase; when the phase becomes imaginary (and the classical dynamics is not relevant anymore), the term $\mathbf{A}_0$ may become relevant. This is another instance of a typically quantum-mechanical effect.

It is also worth noting that the wavefunction ψ (equation (18)) includes momentum $\mathbf{P}=-(q\mathbf{E}_0/\omega)(\cos\omega t-1)$ and energy $\mathcal{E}=-q\mathbf{r}\mathbf{E}_0\sin\omega t$ (from equation (17)); this represents the interaction with "particles" which obey the classical equation of motion $\dot{\mathbf{P}}=-grad\mathcal{E}$.

Unitary transformation. Let us write now

$$\begin{gathered}H_p^{'}=\frac{1}{2m}(\mathbf{p}-\frac{q}{c}\mathbf{A}+\frac{q}{c}\mathbf{A}_0)^2+V(\mathbf{r})=H_0-\frac{q}{mc}(\mathbf{A}-\mathbf{A}_0)\mathbf{p}+\frac{q^2}{2mc^2}(\mathbf{A}-\mathbf{A}_0)^2\ ,\\ i\hbar\frac{\partial\phi}{\partial t}=H_0\phi-\frac{q}{mc}(\cos\omega t-1)\cdot\mathbf{A}_0\mathbf{p}\cdot\phi+\frac{q^2}{2mc^2}A_0^2(\cos\omega t-1)^2\cdot\phi\ .\end{gathered}\tag{23}$$

We introduce now another transformation, given by

$$\begin{gathered}\phi=e^{\frac{iq}{mc\hbar}\int^t dt^{'}(\cos\omega t^{'}-1)\cdot\mathbf{A}_0\mathbf{p}-\frac{iq^2}{2mc^2\hbar}A_0^2\int^t dt^{'}(\cos\omega t^{'}-1)^2}\chi=\\ =e^{-\frac{iq^2}{8mc^2\hbar\omega}A_0^2(\sin 2\omega t-8\sin\omega t+6\omega t)}e^{\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}\chi\ ,\end{gathered}\tag{24}$$

which leads to

$$\begin{gathered}i\hbar\frac{\partial\chi}{\partial t}=e^{-\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}H_0e^{\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}\chi=\\ =\frac{1}{2m}p^2\chi+e^{-\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}V(\mathbf{r})e^{\frac{iq}{mc\hbar\omega}(\sin\omega t-\omega t)\cdot\mathbf{A}_0\mathbf{p}}\chi\ ,\end{gathered}\tag{25}$$

or

$$i\hbar\frac{\partial\chi}{\partial t}=\frac{1}{2m}p^2\chi+\widetilde{V}(\mathbf{r})\chi\ ,\tag{26}$$

where

$$\widetilde{V}(\mathbf{r})=\left[e^{-\frac{q}{m\omega^2}(\sin\omega t-\omega t)\cdot\mathbf{E}_0grad}V(\mathbf{r})\right]\ .\tag{27}$$

The wavefunction ψ is given by

$$\psi=e^{-\frac{iq^2}{8mc^2\hbar\omega}A_0^2(\sin 2\omega t-8\sin\omega t+6\omega t)}e^{-\frac{iq}{\hbar\omega}\mathbf{r}\mathbf{E}_0(\cos\omega t-1)}e^{\frac{iq}{m\hbar\omega^2}(\sin\omega t-\omega t)\mathbf{E}_0\mathbf{p}}\chi\ .\tag{28}$$

Making use of the identity

$$e^Oe^Be^{-O}=e^{[O,B]}e^B\tag{29}$$

for $[O,B]=c\,number$, we put also equation (28) in the form[4]

$$\psi=e^{\frac{iq^2}{8mc^2\hbar\omega}A_0^2(3\sin 2\omega t-8\omega t\cos\omega t+2\omega t)}e^{\frac{iq}{m\hbar\omega^2}(\sin\omega t-\omega t)\mathbf{E}_0\mathbf{p}}e^{-\frac{iq}{\hbar\omega}(\cos\omega t-1)\mathbf{r}\mathbf{E}_0}\chi\ . \tag{30}$$

We can see that in the presence of the electromagnetic radiation the dynamics of the charge is governed by the interaction $\widetilde{V}(\mathbf{r})$. This transformed interaction can be viewed as the structural interaction $V(\mathbf{r})$ (which may be responsible for a bound state) dressed by the radiation. Basically, this dressed interaction is discussed in Refs. [7]-[9].

Let us note, for the sake of the formal completeness, that the above calculations are valid also for an assembly of charges q_a, with mass m_a, $a=1,\,2,....$ The wavefunction is given by

$$\begin{gathered}\psi=e^{-\sum_a\frac{iq_a^2}{8m_ac^2\hbar\omega}A_0^2(\sin 2\omega t-8\sin\omega t+6\omega t)}\times\\ \times e^{-\sum_a\frac{iq_a}{\hbar\omega}\mathbf{r}_a\mathbf{E}_0(\cos\omega t-1)}e^{\sum_a\frac{iq_a}{m_a\hbar\omega^2}(\sin\omega t-\omega t)\mathbf{E}_0\mathbf{p}_a}\chi\ \ ,\end{gathered}\tag{31}$$

where

$$\begin{gathered}i\hbar\frac{\partial\chi}{\partial t}=\sum_a\frac{1}{2m_a}p_a^2\chi+\widetilde{V}(\mathbf{r}_1,\mathbf{r}_2,...)\chi\ ,\\ \widetilde{V}(\mathbf{r}_1,\mathbf{r}_2,...)=\left[e^{-\frac{q_a}{m_a\omega^2}(\sin\omega t-\omega t)\cdot\mathbf{E}_0grad_a}V(\mathbf{r}_1,\mathbf{r}_2,...)\right]\ .\end{gathered}\tag{32}$$

Free charge in radiation field. Let us assume that $V(\mathbf{r})=0$; then $\psi(t=0)=\chi=e^{-\frac{i}{2m\hbar}p^2+\frac{i}{\hbar}\mathbf{pr}}$, where $\mathbf{p}$ (momentum) is a $c-number$. At later times ψ is given by equation (30); we can see that the charge has an average additional momentum $\mathbf{p}_{rad}=q\mathbf{E}_0/\omega$ and an average additional energy $E_{rad}=q^2A_0^2/4mc^2$; we can see that the charge is accelerated rapidly (within the limit $E_{rad}\ll mc^2$). The wavefunctions ψ labelled by $\mathbf{p}$ are orthogonal and complete. We emphasize that these wavefunctions are valid in the region where the radiation is present. The limitation to the factor

$$e^{\frac{iq}{m\hbar\omega^2}\mathbf{E}_0\mathbf{p}\sin\omega t}=\sum_{n=-\infty}^{\infty}J_n(q\mathbf{E}_0\mathbf{p}/m\hbar\omega^2)e^{\frac{i}{\hbar}(n\hbar\omega t)}\tag{33}$$

in the wavefunction ψ (equation (30)), where J_n are the Bessel function of the $n-th$ order, which would imply multiple photons, is not warranted.

Bound charges. Fragmentation (Dissociation). We consider now electrons in atoms, electrons and ions in molecules, in atomic clusters or nucleons in atomic nuclei. Most of these charges, which are relevant for the dynamics, exhibit a quasi-classical motion, due, on one side, to the deep potential wells in heavy atoms and large molecules and, on the other side, to the large ionic mass. We assume that single bound charges are relevant for the dynamics of such assemblies of particles. We assume also that the potential $V(\mathbf{r})$ is responsible for the bound state.

Let us focus on the transformed potential $\widetilde{V}(\mathbf{r})$ of a charge, given by equation (27); it includes the effects the radiation causes on the original potential $V(\mathbf{r})$. The coefficient $\xi=(q/m\omega^2)\mathbf{E}_0grad$ which appears in the exponent of $\widetilde{V}(\mathbf{r})$ can be represented as

$$\xi=\frac{qE_0}{m\omega^2a}=\frac{qA_0}{mc\omega a}=\eta\frac{\lambda}{a}\ \ ,\tag{34}$$

where $\eta=qA_0/mc^2$, a is the size of the assembly along the direction of the electric field and λ is the wavelength of the radiation; for optical radiation λ is of the order $10^{-4}cm$, while for atomic

[4] We note also the identity $e^Oe^B=e^{\frac{1}{2}[O,B]}e^{O+B}$ for $[O,B]$ a $c-number$.

assemblies a is of the order $10^{-8}cm$; we can see that even for a low-intensity radiation ($qA_0/mc^2 \ll 1$) this coefficient is large. The vector potential can be estimated from $A_0 = \sqrt{8\pi cI}/\omega$, where I is the radiation intensity; for $\xi > 1$ and $\eta < 1$ (non-relativistic regime) the intensity I must lie within the range $10^{12}w/cm^2 < I < 10^{18}w/cm^2$ for electrons in atomic assemblies and optical radiation ($\omega = 10^{15}s^{-1}$). Similarly, for protons in atomic nuclei $10^8 w/cm^2 < I < 10^{24}w/cm^2$, while for ions in molecules $10^{18}w/cm^2 < I/A^2 < 10^{24}w/cm^2$, where A is the mass number of the ion. We can see that for protons in atomic nuclei even the available high-intensity radiation remains in the non-relativistic range (and the coefficient given above is very large since a is very small); for ions in molecules in low-intensity radiation the parameter ξ may be smaller than unity. First, let us assume $\xi \gg 1$.

Let us direct the electric field $\mathbf{E}_0$ along the z-direction; from equation (27) we can write

$$\widetilde{V} = V - \frac{qE_0}{m\omega^2}(\sin\omega t - \omega t)V_1 + \frac{q^2E_0^2}{2m^2\omega^4}(\sin\omega t - \omega t)^2 V_2 + ... = V\left(x, y, z - \frac{qE_0}{m\omega^2}(\sin\omega t - \omega t)\right) , \quad (35)$$

where $V_1 = \partial V/\partial z$, $V_2 = \partial^2 V/\partial z^2$; we can see that for a sufficienty large lapse of time the potential $\widetilde{V}$ at the position of the charge is the original potential V at coordinate $z-(qE_0/m\omega^2)(\omega t - \sin\omega t)$; at this point the potential V may vanish; this happens in a time τ (from the application of the radiation field) given by

$$\omega\tau - \sin\omega\tau \simeq \frac{1}{\xi} \ , \quad (36)$$

which leads to

$$\tau \simeq (6/\xi)^{1/3}\omega^{-1} \ . \quad (37)$$

We can see that for $\xi \gg 1$ the time τ is in fact very short; it is much shorter than the radiation period ω^{-1}, which means that during this time the radiation exhibits practically no oscillation. The oscillations appear for much lower radiation intensities ($\xi \ll 1$).

At the moment τ the charge becomes free and the bound state is fragmented (dissociated); the interaction felt by the charge is vanishing and the charge is left with the kinetic energy and momentum acquired while it was localized in a region of the dimension a of the bound state. Therefore, we may take the charge energy of the order $\varepsilon = \hbar^2/ma^2$ and the charge momentum of the order $p = \hbar/a$. In order to escape from the region of the bound state the charge should penetrate a possible potential barrier U. We assume a Coulomb barrier $U = Zq^2/r$, generated by Zq charges, where, for convenience, we introduce the charge number Z. The radial path of the charge ranges from r_1 of the order a up to $r_2 = Zq^2ma^2/\hbar^2$; $a_H = \hbar^2/mq^2$ is the Bohr radius, such that $r_2 = Za^2/a_H$. The relevant factor in the wavefunction χ is $e^{\frac{i}{\hbar}\int_{r_1}^{r_2} dr\cdot p}$, where $p = \sqrt{2m(\varepsilon - U)}$, where we identify the radial momentum with the total momentum (the orbital motion may be left aside); the relevant factors in the total wavefunction ψ given by equation (28) are

$$e^{-\frac{iq}{m\hbar\omega^2}(\omega t - \sin\omega t)E_0\cos\theta\cdot(p_2-p_1) + \frac{i}{\hbar}\int_{r_1}^{r_2} dr\cdot p} \ , \quad (38)$$

where $p_1 = \sqrt{2m(\varepsilon - Zq^2/a)}$, $p_2 = 0$ and θ is the angle between $\mathbf{E}_0$ and $\mathbf{p}$. The transition probability is $w = e^{-\gamma}$, where

$$\gamma = A\xi(\omega t - \sin\omega t)\cos\theta + B$$
$$A = \tfrac{2a}{\hbar}\sqrt{2m(Zq^2/a - \varepsilon)} \ , \ B = \tfrac{2}{\hbar}\int_{r_1}^{r_2} dr\sqrt{2m(Zq^2/r - \varepsilon)} \ ; \quad (39)$$

the time t in this equation is measured from $t = \tau$. We may neglect ε in comparison with Zq^2/a (we assume $Za \gg a_H$) and get

$$\gamma \simeq 2\sqrt{\frac{2Za}{a_H}}\xi(\omega t - \sin\omega t)\cos\theta + \frac{\sqrt{2}a}{a_H}\beta \ , \quad (40)$$

where $\beta = \pi Z$ for $Z \simeq 1$ and $\beta = 2\sqrt{Z}$ for $Z \gg 1$ (in the evaluation of the integral from r_1 to r_2, we have asssumed $a_H \ll Za$). 'The motion proceeds along the path of the minimal phase variation, such that, in the above expression, we have the condition $\gamma > 0$, *i.e.*,

$$\cos\theta > \cos\theta_0 = -\frac{\beta(a/4Za_H)^{1/2}}{\xi(\omega t - \sin\omega t)} \ ; \tag{41}$$

we can see that for $\tau < t < t_0 = (\beta^2 a/4Za_H)^{1/6}\tau$ the ejection of the charge proceeds at all angles, while for $t > t_0 = (\beta^2 a/4Za_H)^{1/2}\tau$ it proceeds only at angles $0 < \theta < \theta_0$; for large times $\theta_0 \to \pi/2$ and the emission proceeds mainly along the direction of the electric field, as expected. The total probability is obtained by integrating over angle θ; we get

$$w_{tot} = \frac{\sqrt{a_H/2Za}}{\xi(\omega t - \sin\omega t)}\sinh\left[2\sqrt{2Za/a_H}\xi(\omega t - \sin\omega t)\right]e^{-\sqrt{2}a\beta/a_H} \ , \ \tau < t < t_0 \tag{42}$$

and

$$w_{tot} = \frac{\sqrt{a_H/2Za}}{2\xi(\omega t - \sin\omega t)}\left[1 - e^{-2\sqrt{2Za/a_H}\xi(\omega t - \sin\omega t) - \sqrt{2}a\beta/a_H}\right] \ , \ t > t_0 \ ; \tag{43}$$

we can see that the probability is dominated by the fast process of dissociation occurring in the short time interval $\tau < t < t_0$ (equation (42)); 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 is their radial velocity (the wavefunction is normalized to the unit volume; actually, it should be normalized to a volume of the order of the atomic dimensions, in accordance with the quasi-classical behaviour of the ejected (free) electron)); 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.

The above calculations may apply to atom ionization in electromagnetic radiation; with a suitable change in the numerical estimations similar calculations can be made for radiation-induced proton emission from atomic nuclei or ion emission from molecules, atomic clusters; in general, by a suitable modification of the charge and mass, such calculations can be performed also for the general process of fragmentation (dissociation) of bound assemblies of charges. The calculations can be extended to multiple ionization, involving also the dynamics of the rest of the assembly.[15]-[17]

The probability w_{tot} (which can be viewed as the transmission coefficient through the potential barrier; we can check that $w_{tot} < 1$) is a function of $\xi(\omega t - \sin\omega t)$; for higher-intensity radiation (increasing ξ) the emission of the charge is faster and in the time interval $\tau < t < t_0 = (\beta^2 a/4Za_H)^{1/6}\tau$ the probability increases (equation (42)).

An interesting question refers to 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 is penetrated very rapidly (practically the barrier may be neglected for high-intensity radiation), and the charge is injected in the high-intensity radiation, where it is rapidly accelerated up to relativistic velocities.[18, 19]

Static field. The above calculations can be applied to a static uniform electric field $\mathbf{E}$ derived from a vector potential $\mathbf{A} = -c\mathbf{E}t$. The wavefunction is

$$\psi = e^{-\frac{iq^2E^2t^3}{6\hbar m}}e^{\frac{iqt}{\hbar}\mathbf{Er}}e^{-\frac{iqt^2}{2\hbar m}\mathbf{Ep}}\chi \ , \tag{44}$$

where χ satisfies the Schrodinger equation

$$i\hbar\frac{\partial\chi}{\partial t}=\frac{1}{2m}p^2\chi+\widetilde{V}(\mathbf{r})\chi \tag{45}$$

with the transformed potential given by

$$\widetilde{V}(\mathbf{r})=\left[e^{\frac{qt^2}{2m}\mathbf{E}grad}V(\mathbf{r})\right] \ . \tag{46}$$

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

$$\tau=\sqrt{\frac{2ma}{qE}} \tag{47}$$

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

$$\gamma=\frac{qEt^2}{m}\sqrt{\frac{2Z}{aa_H}}\cos\theta+\frac{\sqrt{2}a}{a_H}\beta \tag{48}$$

and

$$\cos\theta>\cos\theta_0=-\frac{\beta ma}{qEt^2}\sqrt{\frac{a}{Za_H}} \ . \tag{49}$$

For $\tau<t<t_0=(\beta^2a/4Za_H)^{1/4}\tau$ the emission proceeds in all directions, while for $t>t_0$ it occurs mainly in the forward direction. The total probability is

$$w_{tot}=\frac{m}{qEt^2}\sqrt{\frac{2aa_H}{Z}}\sinh\left(\frac{qEt^2}{m}\sqrt{\frac{2Z}{aa_H}}\right)e^{-\sqrt{2}a\beta/a_H} \ , \ \tau<t<t_0 \tag{50}$$

and

$$w_{tot}\simeq\frac{m}{qEt^2}\sqrt{\frac{aa_H}{2Z}}\left[1-e^{-\frac{qEt^2}{m}\sqrt{\frac{2Z}{aa_H}}-\sqrt{2}a\beta/a_H}\right] \ , \ t>t_0 \ ; \tag{51}$$

the emission time is of the order t_0. For the static field the parameter $\eta=qA/mc^2$ becomes $\eta=\sqrt{2qEa/mc^2}(t/\tau)$.

Low-intensity radiation. If $\xi\ll1$, as in the case of low-intensity radiation, or low static fields (or large dimensions of the assembly), the fragmentation occurs in a long time τ. During this time the radiation field may oscillate many times. The dynamics of the charge is governed by the transformed potential $\widetilde{V}(\mathbf{r})$ given by equation (35). After each time interval τ the charge may "recollide" with the rest of the assembly, and the action of the potential $\widetilde{V}(\mathbf{r})$ is resumed each time.[20]-[22] We can see that the terms of the form $\sin^n\omega t$ in the potential $\widetilde{V}(\mathbf{r})$ generate multiple-photon absorption and, on disexcitation, emission of higher-order harmonics.

Scattering. The wavefunction given by equation (28) is used sometimes for charge scattering; this means that the scattering process is embedded in the radiation, which is not a realistic situation; the asymptotic momenta $\mathbf{p}=m\mathbf{v}+q\mathbf{A}/c$ include the field. In fact, the correct approach would start with the hamiltonian $p^2/2m-q\mathbf{rE}$ and consider the potential $-q\mathbf{rE}$ as the scattering potential (confined to a finite region). Nevertheless, let us consider first $V(\mathbf{r})=0$, *i.e.* scattering off radiation only. The incident charge has the wavefunction $\psi_i\sim e^{i\mathbf{kr}}$ at $z\rightarrow-\infty$ and $t\rightarrow-\infty$ and acquires a wavefunction $\chi\sim e^{i\mathbf{k}_f\mathbf{r}}$ at $z\rightarrow\infty$ and $t\rightarrow\infty$, $k_i=k_f=k$, where $r=(x,y,z)$ and the scattering is concentrated in a region (x,y,z) of the order d (elastic scattering; inelastic scattering is practically inobservable); the order of magnitude of d should be as small as possible, *e.g.* a few radiation wavelengths. The transition probability is given by $|(\psi_i,\psi)|^2$, where ψ is given by equation (28) with χ given here. First we note that in order to see the structure of the scattering the projectile wavelength must be comparable with the radiation wavelength; this makes the projectile to be treated in the quasi-classical approximation, which allows one to use $\mathbf{r}=\mathbf{v}t$ in the transition probability, where $\mathbf{v}$ is the projectile velocity; therefore, we limit ourselves to forward scattering. Similarly, the $\mathbf{r}$-integration may be performed along the trajectory of the projectile with a small cross-section of the order of the projectile wavelength; this means practically an integration over the length l along the trajectory, which permits in fact the use of t instead of l. Then we note that in spite of the smallness of the parameetr ξ the exponents in ψ vary rapidly over the scattering region, which shows that the transition probability is small. The most important contribution comes from $t\sim r\sim l\sim 0$, which makes us to retain only the exponent $\sim \frac{iq}{m\hbar\omega^2}\sin\omega t\cdot\mathbf{E}_0\mathbf{p}$ (where $\mathbf{p}$ is $\hbar\mathbf{k}_f$); here we may use the expansion given by equation (33); we get diffraction maxima at scattering angles given by $k\theta_n=\omega n/v$, where v is the projectile velocity; the amplitudes of the transition probability are proportional to $J_n(dk\xi)$. The diffraction maxima require emission of photons, which restricts the labels n to positive values. This scattering process amounts to an infinite summation of the Born approximation in all orders.

If an additional scattering potential $V(\mathbf{r})$ is present, it is easy to see that the wavefunction χ is proportional to $\chi\sim e^{i\mathbf{k}_f\mathbf{r}}f$, where f is the scattering amplitude due to the potential $V(\mathbf{r})$; it follows that the cross-section for a given diffraction maximum is given by $|J_n|^2\sigma_V$, where σ_V is the cross-section due to the potential $V(\mathbf{r})$; this is known as the Kroll-Watson formula.[23] We emphasize that the conservation laws of momentum (and energy for inelastic scattering) are very approximately satisfied in fact, due to the restricton of the range of integration. Moreover, the emission-photons scattering would mean an unphysical spontaneous release of energy.

Concluding remarks. The "dipole" hamiltonian and the "standard" non-relativistic hamiltonian of charges in electromagnetic radiation are derived from the mechanical action. Making use of suitable unitary transformations of the Kramers-Henninger type, it is shown that they are equivalent. A unitary transformation is introduced which takes into account the modifications brought about by the electromagnetic radiation upon the original interaction of the charges (dressed interaction). It is shown that the radiation favours the fragmentation of the bound states of charges, setting them free to penetrate a possible potential barrier. Explicit calculations are presented for a Coulomb potential barrier and the emission probability and emission time are calculated. Applications are discussed to ionization of atoms and molecules, radiation-induced proton emission from atomic nuclei, ion emission from large molecules or fragmentation of atomic clusters.

Finally, it is worth noting the peculiar character of the "quasi-classical" tunneling, used to estimate the probability emission of a charge from a bound state.[5, 6] In the "quasi-classical" tunneling the penetration of the potential barrier proceeds when the interaction energy $(\mathbf{p}-q\mathbf{A}/c)^2/2m$ with $\mathbf{p}=0$ equals the binding energy $-E_b$ ($E_b>0$); this happens at imaginary times. For instance, for a static field $q^2E^2t_i^2/2m=-E_b$, hence $t_i=i(2mE_b)^{1/2}/qE$. The wavefunction acquires the factor $e^{-\frac{i}{\hbar}\int^{t_i}dt(q^2E^2t^2/2m)}$, hence the decay probability $e^{-(2mE_b)^{3/2}/3\hbar mqE}$, with the characteristic $1/E$-dependence in the exponent; the quasi-classical behaviour is valid for small E (small probability). This is a crude approximation which neglect essential features of the dynamics.

A related procedure for estimating such probabilities in static fields is the following. The energy $-\mathbf{r}\mathbf{E}$ is viewed as a potential barrier for the bound-state energy $-E_b$; then, the momentum is $p=i\sqrt{2m(-xE+E_b)}$, where x denotes the direction along the field; the evaluation of the integral $-(2/\hbar)\int_0^{E_b/E}dxp$ gives the exponent $\sim const/E$. The difficulty is that this is not a tunneling problem, since for $x\rightarrow-\infty$ the momentum $p=\sqrt{2m(xE-E_b)}$ is not real (similarly, the imaginary-time tunneling described above is not a tunneling problem for bound charges); it is a problem involving the charge motion in the electric field. If the electric field is sufficiently strong we may neglect the atomic interaction, and the solution is a wavefunction ψ_f of the Airy type, as it is well known. We may estimate the transition probability from the original atomic state described by a wavefunction ψ_b (corresponding to a bound state) to the final state described by ψ_f (practically a free, extended state) by $|(\psi_f, \psi_b)^2|$. If the interaction is switched on slowly (adiabatically), there is no transition; if the interaction is switched on rapidly, there are transitions; usually, their probabilities are small, since the scalar product is between a very localized state and a very extended one. In any case, this is not the problem we deal with in the case of laser-matter interaction, where we are interested in transition probabilities produced at finite times by an oscillating, or constant, field.

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