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A note on the ionization of atoms by electromagnetic radiation

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Abstract

The Goeppert-Mayer and Kramers-Henneberger transformations are examined for bound charges placed in electromagnetic radiation in the non-relativistic approximation. The consistent inclusion of the interaction with the radiation field provides the time evolution of the wavefunction with both structural interaction (which ensures the bound state) and the interaction caused by the radiation. It is shown that after a short time since switching on a high-intensity radiation the bound charges are set free; in these conditions a statistical criterion is used to estimate the rate of atom ionization. The results correspond to a sudden application of the interaction with the radiation, in contrast with well-known ionization probability obtained by quasi-classical tunneling through classically unavailable non-stationary states, or other equivalent methods, where the interaction is applied adiabatically. For low-intensity radiation the charges oscillate and emit higher-order harmonics, the charge configuration is rearranged and the process is resumed. 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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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 the presence of the radiation implies a quasi-classical tunneling through states which are not allowed by the classical dynamics (including imaginary time tunneling[4]). Later, it was realized[5] that the radiation-dressed states play an important role in the ionization process through the Kramers-Henneberger transformation.[6, 7] Similarly, the ionization rate in static electric field was computed in classical works,[8]-[10] either by quantum transitions or by tunneling through the potential barrier generated by the field. These calculations assume an adiabatic introduction of the interaction with the radiation, which allows the use of atomic stationary states.[11] Typically, the dynamics of the electrons in the presence of the radiation field is neglected in these approaches, which results in ionization probabilities proportional to $e^{-const/E}$, where E is the strength of the electric field[12]-[15] (this is a well-known result, valid also for static fields[4]). In current experiments envisaging atom ionization, especially in high-intensity electromagnetic radiation, the interaction occurs suddenly in the focal region of the laser beam. In this case, the atomic states are not stationary anymore, and a time evolution of the wavefunction is needed. On a sudden application

of the interaction, both in static and time-dependent electromagnetic field, the particle energy is not determined. The need of a time-evolving picture of radiation-induced atom ionization has often been emphasized.[16]-[18] We present here an investigation into the time-evolution of bound states of charges for a sudden application of the interaction with the electromagnetic radiation.

First, we show that the consistent inclusion of the interaction with the radiation field by means of the unitary transformations of the Goeppert-Mayer and Kramers-Henneberger type offers the opportunity to follow the time evolution of the wavefunction with both the structural interaction (which is responsible for the charge bound state) and the interaction caused by the radiation. Second, it is shown that in high-intensity radiation, after a short lapse of time since its application, the well-known radiation-dressed atomic interaction resulting from the Kramers-Henneberger transformations vanishes, setting the electrons free; using a statistical criterion we derive the atom ionization rate, which differs from the known results obtained by introducing the interaction adiabatically. The effect of low-intensity radiation is that of an adiabatic perturbation. During its slow action, the electrons oscillate among stationary states, leading to well-known multi-photon absorption and production of high-order electromagnetic harmonics, the rearrangement of the charge configuration, the process being periodically resumed. We apply the results of this new context to the ionization of atoms, assuming, for convenience, (non-relativistic) single-electron states in atomic mean-field. The extension of the results to multiply-charged ions, ionization of large molecules, atomic clusters, or 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 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 $\mathbf{E} = \mathbf{E}_0 \sin(\omega t + \alpha)$, where ω is the radiation frequency (linear polarization), t denotes the time and α is an initial phase. We consider the associated Schrodinger equation $i\hbar\partial\psi/\partial t = H_d\psi$ and introduce the unitary transformation

$$\psi = e^{iS_1}\phi \ , \ \ S_1 = -\frac{q}{\hbar\omega}\mathbf{r}\mathbf{E}_0 \left[\cos(\omega t + \alpha) - \cos\alpha\right] \ \ , \tag{2}$$

where we recognize the vector potential $\mathbf{A} = (c/\omega)\mathbf{E}_0 \left[\cos(\omega t + \alpha) - \cos\alpha\right]$ ($\mathbf{E} = -(1/c)\partial\mathbf{A}/\partial t$), $\hbar$ being the Planck's constant and c the speed of light in vacuum; we can write $S_1 = -(q/\hbar c)\mathbf{r}\mathbf{A}$. The transformation given by equation (2) leads to $\mathbf{p} \to \widetilde{\mathbf{p}} = \mathbf{p} - q\mathbf{A}/c$ and the standard non-relativistic hamiltonian

$$\widetilde{H}_d = e^{-iS_1} H_d e^{iS_1} = \frac{1}{2m}\left(\mathbf{p} - \frac{q}{c}\mathbf{A}\right)^2 + V(\mathbf{r}) \ \ , \tag{3}$$

with the associated Schrodinger equation $i\hbar\partial\phi/\partial t = \widetilde{H}_d\phi$. The transformation given by equation (2) is known as the Goeppert-Mayer transformation.[19]

Let us write

$$\widetilde{H}_d = H_0 - \frac{q}{mc}\mathbf{A}\mathbf{p} + \frac{q^2}{2mc^2}A^2 \tag{4}$$

and continue with the unitary transformation

$$\phi = e^{iS_2} e^{iS_3} \chi \ ,$$

$$S_2 = -\frac{q^2}{2\hbar m c^2} \int_0^t dt A^2 = -\frac{q^2 E_0^2}{8\hbar m \omega^3} [\sin 2(\omega t + \alpha) - \\ -8\cos\alpha \cos(\omega t + \alpha) + 2\omega t(2 + \cos 2\alpha) + 3\sin 2\alpha] \ , \tag{5}$$

$$S_3 = \frac{q}{\hbar m c} \int_0^t dt \mathbf{pA} = \frac{q}{\hbar m \omega^2} \left[\sin(\omega t + \alpha) - \omega t \cos\alpha - \sin\alpha\right] \mathbf{E}_0 \mathbf{p} \ ;$$

it leads to the Schrodinger equation

$$i\hbar \frac{\partial \chi}{\partial t} = \left[\frac{1}{2m} p^2 + \widetilde{V}(\mathbf{r})\right] \chi \tag{6}$$

with the radiation-dressed atomic potential

$$\widetilde{V}(\mathbf{r}) = e^{-\frac{q}{m\omega^2}[\sin(\omega t + \alpha) - \omega t \cos\alpha - \sin\alpha] \cdot \mathbf{E}_0 grad} V(\mathbf{r}) \ ; \tag{7}$$

we note that the interaction with the radiation is applied suddenly at $t = 0$, where $\psi(t = 0) = \phi(t = 0) = \chi(t = 0)$ and the electromagnetic interaction vanishes at $t = 0$ in the standard non-relativistic hamiltonian given by equation (4) and in $\widetilde{V}(\mathbf{r})$; this establishes the equivalence of the three hamiltonians (equations (1), (4) and (6)) and the consistent inclusion of the interaction with the radiation field (see, for instance, Refs. [20]-[22]). The unitary transformations given by $S_{2,3}$ are the well-known Kramers-Henneberger transformations, including the radiation-dressed potential $\widetilde{V}(\mathbf{r})$.[5]-[7] The wavefunction $\psi = \exp(iS_1)\exp(iS_2)\exp(iS_3)\chi$ given by the above formulae is known as the non-relativistic Volkov wavefunction;[18] its expansion in a temporal Fourier series indicates the presence of multiple "photons" with frequencies $n\hbar\omega$, where $n = 0, 1, 2, ...$ integer.[14, 23, 24]

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 (7) gives

$$\widetilde{V}(x, y, z) = V + \zeta(t) V_1 + \tfrac{1}{2!}\zeta^2(t) V_2 + ... = \\ = V\left(x, y, z + \zeta(t)\right) \quad , \tag{8}$$

where $V_1 = \partial V/\partial z$, $V_2 = \partial^2 V/\partial z^2$, ... and

$$\zeta(t) = \frac{qE_0}{m\omega^2} \left[\omega t \cos\alpha - \sin(\omega t + \alpha) + \sin\alpha\right] \ . \tag{9}$$

We can see that the potential $\widetilde{V}$ at the position of the charge is the original potential V at coordinate $z + \zeta(t)$. It is convenient to introduce the parameter $\xi = |q|\, E_0/m\omega^2 a$, where a denotes a length of the order of the dimension of the atom (in its ground-state); we asssume $a \simeq a_H = \hbar^2/me^2$, where a_H is the Bohr radius,$q = -e$ being the electron charge.

For $\xi \ll 1$ (low-intensity radiation) the electronic charge oscillates and emits higher-order harmonics, due to the oscillations brought about by $\zeta(t)$ in the z-coordinate; during this process the charge is reconfigured, the time of charge re-arrangement being given approximately by $t_a = \hbar/\Delta\mathcal{E}$, where $\Delta\mathcal{E}$ is the perturbation energy generated by the radiation field via the potential $\widetilde{V}$. During the

time t_a the potential V changes and its radiation-dressing process is resumed periodically. This is a well-known process.[25]-[28]

For $\xi \gg 1$ (high-intensity radiation) the deviation $\zeta(t)$ may get rapidly larger than the atom size, and the electrons are set free. The attractive potential of the resulting ion is surpassed by the kinetic energy of the ejected electron. The parameter ξ can also be written as $\xi = \eta(\lambda/a)$, where $\eta = |q|\, A_0/mc^2$, $A_0 = cE_0/\omega$, λ being the radiation wavelength. In order to preserve the non-relativistic approximation we should have $\eta \ll 1$. The conditions $\xi > 1$ and $\eta < 1$ are satisfied for radiation intensity I in the range $10^{11} w/cm^2 < I < 10^{18} w/cm^2$ for electrons in atoms ($a = 10^{-8} cm$) and optical radiation ($\omega = 10^{15} s^{-1}$, $\lambda \simeq 10^{-4} cm$). Even for moderate laser intensities the parameter ξ is large ($\xi \simeq 10^3$ for $I = 10^{18} w/cm^2$). Under these circumstances the duration τ for setting the electrons free is given by

$$|\zeta(\tau)|\,/a = \xi\,|\omega\tau\cos\alpha - \sin(\omega\tau+\alpha) + \sin\alpha| \simeq \frac{1}{2}\xi(\omega\tau)^2\,|\sin\alpha| = 1\ . \tag{10}$$

We can see that the displacement of the electrons is in the direction opposite to the electric field at the initial moment, as expected. Depending on the sign of $\sin\alpha$, this displacement is either positive or negative. We assume the phase α randomly distributed and take the mean value $\overline{|\sin\alpha|} = 2/\pi$; we use $\xi(\omega\tau)^2/\pi = 1$ as a statistical criterion for ionization.

Let us consider an atom with the nucleus charge Ze; according to the Thomas-Fermi model for heavy atoms ($Z \gg 1$) the electrons are concentrated mainly up to a distance of the order $a_H/Z^{1/3}$ from the nucleus; therefore, we may estimate the release time τ from $\xi(\omega\tau)^2/\pi = 1 - 1/Z^{1/3}$. For an ion with one electron and the nuclear charge Ze the radius of the electron orbit is of the order a_H/Z^2 and the time τ can be estimated from $\xi(\omega\tau)^2/\pi = 1 - 1/Z^2$. In general, for a heavy ion with charge ne, $n \ll Z$, the charge localization distance b can be estimated from $Ze^2/b \simeq \hbar^2(Z-n)^{2/3}/mb^2$, which gives $b \simeq a_H(Z-n)^{2/3}/Z$. In all these cases, for large Z, we may take approximately the condition $\xi(\omega\tau)^2/\pi \simeq 1$ for estimating the release time τ. Similary, the same condition is valid for light atoms. In these conditions, the statistical criterion of ionization makes no distinction between successive ionization acts or multiple ionization, the very short release time being the same for each electron. After each process of electron ejection the electronic core suffers a reconfiguration (re-arrangement) process and the potential $V(\mathbf{r})$ is modified; this is the well-known process of core "shake-up" (or core excitation).[27]-[30] As a consequence of this reconfiguration process, the condition of setting the electrons free derived from equation (8) is not valid anymore; a new bound state is formed and a new transformation process begins for the modified potential $V(\mathbf{r})$. For high-intensity radiation the re-arrangement time is comparable with the release time τ, such that we may estimate the ionization rate as

$$\frac{1}{\tau} \simeq \sqrt{\xi/\pi}\,\omega = \sqrt{qE_0/\pi ma}\ ; \tag{11}$$

the decay law of atom population N is $N = N_0 e^{-t/\tau}$, where N_0 is the initial number of atoms. We can see from equation (11) that the uncertainty in energy brought about by the ionization is $\Delta\mathcal{E} = \hbar/\tau \simeq \sqrt{(\hbar^2/ma^2)qE_0a}$; the mechanical work qE_0a done by the field to extract the electron is of the same order of magnitude as the localization energy $\hbar^2/ma^2$ for atomic fields $E_0 \simeq 10^6 statvolt/cm$ (intensity $I \simeq 10^{14} w/cm^2$), as expected; in this case $\xi = (\hbar^2/ma^2)^2/(\hbar\omega)^2 = 10^2$ and $1/\tau = [(\hbar^2/ma^2)/\pi(\hbar\omega)]\,\omega = (10/\pi)\omega$ (it is assumed that the laser pulse duration is longer than the time τ). For $E > E_0$ the process of interaction with the radiation is dominated by ionization. For weaker fields the ionization process slows down appreciably for mechanical work qE_0a smaller than $-\mathcal{E}_b$, where $\mathcal{E}_b$ (< 0) is the binding energy of the electron.

We can get a critical value E_{0c} of the electric field for the transition from the low- to high-intensity regime from $\xi = 1$; this condition gives $E_{0c} = m\omega^2 a/q$. For electrons ($q = 4.8 \times 10^{-10} esu$), optical

radiation $\omega = 10^{15}s^{-1}$ and $a = 10^{-8}cm$ we get $E_{0c} \simeq 2 \times 10^4 statvolt/cm$; this field corresponds to a radiation intensity $I \simeq 10^{11}w/cm^2$. We can see from equation (11) that the ionization rate increases with increasing field strength. In the intermediate-intensity range corresponding to $\xi \simeq 1$ the interplay between highly-excited states and ionization may generate a transient regime of atomic stabilization, with a lower ionization rate.[31] In this region the ionization rate $1/\tau$ can be estimated by solving the equation $|\zeta(\tau)|/a = 1$ for τ; it is easy to see that the solution $\tau(E)$ exhibits oscillations with respect to the field strength E.

We note that the above calculations are done for linearly polarized radiation; it is easy to see that similar calculations can be done for a general polarization.

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 charge being rapidly injected in the high-intensity radiation, where it is accelerated up to relativistic velocities.[32]-[34]

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

$$\psi = e^{-\frac{iq^2E^2t^3}{6\hbar m}} e^{\frac{iqt}{\hbar}\mathbf{Er}} e^{-\frac{iqt^2}{2\hbar m}\mathbf{Ep}} \chi \quad , \tag{12}$$

where χ satisfies the Schrodinger equation

$$i\hbar \frac{\partial \chi}{\partial t} = \frac{1}{2m} p^2 \chi + \widetilde{V}(\mathbf{r})\chi \tag{13}$$

with the transformed potential given by

$$\widetilde{V}(\mathbf{r}) = e^{\frac{qt^2}{2m}\mathbf{E}grad} V(\mathbf{r}) \ . \tag{14}$$

It is easy to see that the ionization rate is $1/\tau = (qE/2ma)^{1/2}$; it coincides practically with the ionization rate in oscillating fields (equation (11)).

Finally we note that the displacement occurring in the radiation-dressed interaction (equation (8)) is given, in general, by

$$\boldsymbol{\zeta}(t) = \frac{q}{m} \int_0^t dt_1 \int_0^{t_1} dt_2 \mathbf{E}(t_2) \ ;$$

by using this equation we can estimate the ionization rate for a general time-dependence of the electric field in the laser pulse. For example, for a very short δ-like pulse $\mathbf{E} = T\mathbf{E}_0\delta(t - t_0)$ occurring at t_0 with a width T we get a ionization rate $1/\tau \simeq qTE_0/ma$; this equation differs from the $\sqrt{E_0}$-dependence given above. Since strong fields are obtained usually with very short pulses, the linear dependence on the field indicated here may be more suitable for analyzing experimental data in this case.[35]

In conclusion, we may say that by means of suitable unitary transformations of the Goeppert-Mayer and Kramers-Henneberger type the interaction of bound charges with the electromagnetic radiation can be consistently taken into account for a sudden application of the interaction with the radiation. It is shown that the radiation-dressing of the structural interaction (which is responsible of the charge bound state) favours, in this case, the dissociation of the bound state (ionization)

in high-intensity radiation; the atom ionization rate is estimated by using a statistical criterion. The new picture of atomic ionization described in this paper implies an explicit time evolution of the charge wavefunction with both structural and radiation interaction; in a short time τ after turning-on the high-intensity radiation the bound charges are set, practically, free.

The approach presented here can be extended to the ionization of molecules[36, 37] or atomic clusters,[38] or to proton emission from atomic nuclei,[39] or even ion emission from molecules (atomic clusters). For electrons in atoms the conditions of ionization and non-relativistic approximation imply a radiation intensity in the range $10^{11}w/cm^2 < I < 10^{18}w/cm^2$, for atomic dimensions of the order $a = 10^{-8}cm$ and optical radiation with frequency $\omega = 10^{15}s^{-1}$; similar conditions lead to $10^{7}w/cm^2 < I < 10^{23}w/cm^2$ for proton emission from atomic nuclei and $10^{17}w/cm^2 < I/A^2 < 10^{23}w/cm^2$ for ion emission from molecules, where A is the mass number of the ion.

A note on the difference between adiabatic application of the interaction and its sudden application. The radiation wavefront is extending much into space, in advance of the front, by the macroscopic process of creating the wave (*e.g.*, by opening the aperture of the laser cavity for releasing the radiation beam, or by switching on the electric circuit of lighting up the lamp, etc); the radiatin wavefront is not sharp. As such, the particle suffers first a very small interaction, which increases gradually in long time. Strictly speaking, the first impact with the radiation is sudden, and, therefore, an uncertainty in energy occurs in a very short time, of the order a/c. However, if there exists energy loss (as in case of condensed matter), this time is longer, and we may consider the interaction application as being adiabatic; it is expressed by the characteristic factor $e^{-\alpha t}$ in the interaction. If we inject (inevitably) slowly non-relativistic charges into the focal region of the laser beam, we introduce again adiabatically the interaction. But, if the laser beam hits the charge, the interaction is applied suddenly. This sudden application of the interaction (which is frequent, for example, in gases), if weak, gives a transient regime, which fads out in time as a consequence of the re-arrangement process; such that, in the stationary regime which follows we may again admit that the interaction is introduced adiabatically. If the interaction is strong, it is the transient regime which dominates.

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