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NON-PERTURBATIVE METHODS APPLIED TO MULTIPHOTON IONIZATION*

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ABSTRACT. ~~We discuss~~ The use of non-perturbative methods in the treatment of atomic ionization. Particular attention is given to schemes of the type proposed by Keldysh where multiphoton ionization and tunnel auto-ionization occur for high intensity fields. These methods are shown to correspond to a certain type of expansion of the T-matrix in the intra-atomic potential; in this manner a criterium concerning the range of application of these non-perturbative schemes is suggested. A brief comparison between the ionization rate of atoms in the presence of linearly and circularly polarized light is presented.

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RESUMO. ~~Discutimos~~ o uso de métodos não perturbativos no tratamento da ionização de átomos. Atenção particular é devotada a modelos do tipo proposto por Keldysh, onde ionização por muitos fótons e auto-ionização por efeito tunel ocorrem para campos muito intensos. Mostra-se que estes métodos correspondem a um certo tipo de expansão da matriz T no potencial intra-atômico e um critério, para o intervalo onde são válidos estes modelos, é sugerido. É apresentada, de forma resumida, uma comparação entre as razões de ionização para átomos em presença de luz linearmente e circularmente polarizada.

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1. INTRODUCTION

The existence of modern lasers available in frequency ranges varying from the far infrared up to the vacuum-ultraviolet region, as well as focused pulsed laser beams with peak power densities above gigawatts per cm^2 , greatly stimulated during the past decade the investigation of the physics of atoms, molecules and solids in the presence of strong coherent radiation fields. Particular attention has been given, both theoretically and experimentally, to the study of particle scattering, atomic excitation, atomic ionization, and molecule dissociation in the presence of intense electromagnetic fields (EMF). These subjects have been reviewed by several authors and we mention as references the works of Delone [1], Lambropoulos [2], Bayfield [3] and Ehlötzky [4]. For resonance phenomena, new effects occur at moderate laser intensities. For instance under adequate conditions the Weisskopf-Wigner Lorentzian lineshape gets altered for intensities of the order of $1\text{W}/\text{cm}^2$, and two extra peaks show up in the spectra constituting the so-called dynamical Stark effect.

In this work we discuss atomic ionization in the presence of strong EMF when resonance processes are not involved. In this case, non-linear effects appear at high laser intensities, which are experimentally attained by CO_2 and Neodimium lasers. Non-linearity implies that multiphoton processes become significant as compared to one photon absorption; the theoretical description of the phenomena occurring under such condition lies outside the realm of lowest order perturbation theory and has

challenged the theoreticians during the past decade. The theoretical treatments of multiphoton ionization can be roughly divided in two classes:

a) Perturbation theory in the atom-laser coupling Hamiltonian (H_I). The transition rate associated to n-photon processes are obtained by evaluating the n-th order contribution to the evolution operator (or equivalently to the T-matrix):

$$U^{(n)}(t) = (i)^{-n} \int_0^t dt_n \int_0^{t_n} dt_{n-1} \dots \int_0^{t_2} dt_1 H_I(t_n) H_I(t_{n-1}) \dots H_I(t_1)$$

The transition rate for a n-th order process is proportional to E_0^{2n} , where E_0 is the EMF amplitude, as long as no intermediate resonances occur.

Therefore higher order perturbation theory predicts that a log-log plot of the ionization rate (or ionization probability) versus the intensity of the laser field is a straight line, with slope equal to the multiplicity of the process (n). In fact this has been verified by Lompre et al [5] for field intensities up to 10^{15} W/cm². On the other hand, recently, Boreham et al [6] have reported deviations from the predictions of perturbation theory for field intensities of the order of 10^{16} W/cm². For very intense fields, perturbation theory becomes questionable due to convergence problems. Alternative approaches to perturbation theory have been suggested by several authors and they lie in the class of the usually called "non-perturbative" methods.

b) "Non-perturbative" methods, generally denominate the approaches which attempt to include the EMF to all orders

in an approximated manner.

The present work concerns a particular type of "non-perturbative" treatment which originated with the pioneer work of Keldysh [7], almost two decades ago, and whose expression for the ionization probability fits the experimental results of Boreham et al [6] and Baldwin and Boreham [8]. Many have been the works based in approaches similar to Keldysh's, making use of the fact that the Schrödinger equation for an electron in the presence of an oscillating, monochromatic EMP can be solved exactly (originating the so-called Volkov solutions [9]). These approximations we denominated high intensity approximations. Without being exhaustive, we mention the works of Perelomov et al [10], the relativistic approach for spin zero particles of Nikishov and Ritus [11], the works closely related to these two of Berson [12], Manakov and Rapoport [13], Geltman and Teague [14], Gersten and Mittleman [15] and of Pert [16]; the space translation approximation (STA) as proposed by Faisal [17] for ionization, etc... Recently Brandi and Davidovich [18] and Brandi et al [19] have shown that some of the most commonly used high-intensity approximations, namely the Keldysh approximation [7], the Perelomov et al. approach [10], the Gersten and Mittleman formulation [15] and the STA [17] correspond to the same kind of expansion of the Green's function in the intra atomic potential and differ at most by a unitary or by a gauge transformation. Although many of the previous mentioned approaches shed light in the understanding of the relation between multiphoton and tunneling processes as mechanisms for atomic ionization, it still remained the question concerning

their range of validity. This is one of the questions we discuss in the following sections, together with the ionization rate of atoms by strong linearly and circularly polarized laser light.

II. IONIZATION IN AN ALTERNATING ELECTRIC FIELD AND THE ADIABATIC APPROXIMATION

In order to have better understanding of the ionization process occurring in the presence of an alternating EMF we firstly discuss a simpler problem: atomic ionization in the presence of a static electric field $\vec{E} = E\vec{z}$. In this case the Hamiltonian is given by (atomic units are used throughout this work)

$$H = \frac{P^2}{2} + V(\vec{r}) - Ez \quad (1)$$

where $V(\vec{r})$ is the intra atomic potential. The electron initially bound with energy $-I_0$ can ionize by tunnelling. The ionization rate, for fields not very strong, may be calculated by the WKB method [20], and for the hydrogen atom initially in the ground state it yields

$$W_{St.} = 8I_0 \left(\frac{E_0}{E}\right) \exp\left(-\frac{2}{3} \frac{E_0}{E}\right) \quad (2)$$

where E_0 is the amplitude of the electric field due to the atomic nucleus in the first Bohr orbit $E_0 = (2I_0)^{3/2} = 5 \times 10^9$ V/cm.

Eq.(2) is valid for $E \ll E_0$.

A similar situation occurs for an alternating electric field if the period of the field $T_F = \frac{2\pi}{\omega}$ is much larger

than the ionization time T_I , which can be estimated from Eq. (2) as

$$T_I \approx \frac{1}{W_{St}} = \frac{1}{8I_0} \left(\frac{E}{E_0}\right) \exp\left(\frac{2}{3} \frac{E_0}{E}\right) \quad (3)$$

Therefore

$$\frac{T_I}{T_F} = \frac{\gamma}{8\pi} F(E/E_0) \quad (4)$$

where we have introduced the quantities $\gamma \equiv \frac{1}{2}(\omega/I_0)(E_0/E)$ and $F(X) = X^2 \exp(-\frac{2}{3X})$.

For fields such that $T_I/T_F \ll 1$, the ionization rate may be obtained by averaging Eq. (2) over one cycle of the alternating field (of course for the WKB to be valid $E \ll E_0$), yielding [10] $W_{ad} = (3E/\pi E_0)^{1/2} W_{St}$. From Eq. (4) it is clear that $T_I/T_F \ll 1$ is satisfied if $\gamma \ll 1$ (in this connection γ has been called the adiabaticity parameter). However this is not a sufficient condition to have $T_I/T_F \ll 1$, since the ratio E/E_0 is also present in Eq. (4). For typical values of, $I_0 = 10\text{eV}$ (~ 0.37 a.u.) $\omega = 1\text{eV}$ (~ 0.037 a.u.), one should have $E > E_0/10$ in order to satisfy $T_I/T_F \ll 1$. But for these values of the field, the application of the WKB method may be questionable.

III. THE HIGH INTENSITY APPROXIMATION

An alternative approach to the one discussed in Sec. II has been proposed by Keldysh [7] and it contains essentially the whole idea behind the high-intensity approximations. Starting from the transition amplitude (Keldysh [7], STA [17])

$$T_{fi} = \int_{-\infty}^t dt' \langle \psi_f, H_I \phi_0 \rangle \quad (5)$$

where, $\langle \rangle$ denotes integration over space coordinates, ψ_f is a Volkov state [9], ϕ_0 is the atomic bound state in the absence of EMF and H_I is the laser atom coupling Hamiltonian, Brandi and Davidovich [18] have shown that Eq.(5) can be expressed as the first terms of an expansion in powers of the intra atomic potential $V(|\vec{x}|)$. In fact, let us introduce the Green's functions G'_0 and G defined by

$$\left[i \frac{\partial}{\partial t} - \frac{p^2}{2} - H_I(t) \right] G'_0(\vec{x}t, \vec{x}'t') = \delta^3(\vec{x}-\vec{x}') \delta(t-t') \quad (6)$$

and

$$\left[i \frac{\partial}{\partial t} - \frac{p^2}{2} - H_I(t) - V(|\vec{x}|) \right] G(\vec{x}t, \vec{x}'t') = \delta^3(\vec{x}-\vec{x}') \delta(t-t'). \quad (7)$$

The Volkov solutions can be written in terms of the Green's function G'_0 defined by Eq.(6) as

$$\psi_f(\vec{x}, t) = \phi_f(\vec{x}, t) + \int d^3x' \int_{-\infty}^t dt' G'_0(\vec{x}t, \vec{x}'t') H_I(t') \phi_f(\vec{x}'t') \quad (8)$$

where ϕ_f is a plane wave.

Of course the Hamiltonian H_I depends on the choice of gauge. If we substitute Eq.(8) into (5) we may rewrite it symbolically as

$$T_{fi} = \langle \phi_f | H_I | \phi_0 \rangle + \langle \phi_f | H_I G'_0 H_I | \phi_0 \rangle \quad (5.a)$$

where the symbol $\langle || \rangle$ stands for integration over all time and space coordinates. This expression is the same used by Gersten and Mittleman [15] except that they neglect terms

originating from $A^2/2c^2$ in the coupling Hamiltonian, Eq.(5.a) may be compared with the exact transition matrix which is expressed in terms of the total Green's function (7) as

$$T_{fi}^{EX} = \langle \phi_f | H_I | \phi_0 \rangle + \langle \phi_f | H_I G H_I | \phi_0 \rangle , \quad (9)$$

if one uses the relation

$$G(\vec{x}t, \vec{x}'t') = G_0^+(\vec{x}t, \vec{x}'t') + \int d^3x'' dt'' G_0^+(\vec{x}t, \vec{x}''t'') V(|\vec{x}''|) G(\vec{x}''t'', \vec{x}'t') \quad (10)$$

Eq.(5.a) may be regarded as the first two terms of an expansion of the transition matrix in terms of the intra atomic potential V , which is equivalent to replacing in the exact T matrix (Eq.(9)) G by G^+ . This approximation is exact in the limiting situations where $V \rightarrow 0$ or $E \rightarrow \infty$, and it is also evident from the first term on the right hand side of Eq.(5.a) that first order perturbation theory is recovered by this type of approximation.

A criterium concerning the range of validity of the high-intensity approximations can be established by considering the first term of the T matrix depending on the intra atomic potential (V), and imposing a condition for fast convergence of the series expansion [18] i.e.

$$\frac{\langle \phi_f | H_I G_0^+ V G_0^+ H_I | \phi_0 \rangle}{T_{fi}} \ll 1 . \quad (11)$$

An exact evaluation of the numerator of inequality (11) is complicated, but approximate calculations have shown that the above condition is satisfied if the adiabaticity parameter is small as compared to unity (in fact within the approximations

considered inequality (11) is satisfied for $\gamma^6 \ll 1$).

To evaluate (6) we consider for convenience [19] the gauge $\phi=0$, $\text{div}\vec{A}=0$, so that

$$H_I(t) = -\frac{1}{2} \vec{p} \cdot \vec{A}(t) + \frac{1}{2c^2} A^2(t). \quad (12)$$

For linearly polarized fields with

$$\vec{A}(t) = \frac{c\vec{E}}{\omega} \cos\omega t; \quad \vec{E} = E\hat{x} \quad (13)$$

it can be shown [19] that the multiphoton transition rate from the initial state ϕ_0 to a final state with momentum \vec{p} is given by

$$W_{fi}^{(N)} = \frac{|\tilde{\phi}_0(\vec{p})|^2}{(2\pi)^2} \left(I_0 + \frac{E^2}{2}\right)^2 \left[\sum_{n=-\infty}^{\infty} J_{N+2n}\left(\frac{\vec{p}\cdot\vec{E}}{\omega^2 c}\right) J_n\left(\frac{E^2}{8\omega^3 c}\right) \right]^2 \delta\left(\frac{p^2}{2} + I_0 + \frac{E^2}{4\omega^2 c^2} - N\omega\right) \quad (14)$$

where $\tilde{\phi}_0(\vec{p})$ is the Fourier transform of $\phi_0(\vec{R})$.

The ionization threshold is $I_0 + E^2/4\omega^2 c^2 = I_0(1 + 1/2\gamma^2)$, instead of I_0 . The extra term $I_0/2\gamma^2$ has been discussed by Kibble [21], who has shown that it can be interpreted as a potential energy associated with the ponderomotive force, when the motion of the electron is averaged over many periods of the applied field. As the electron leaves the focal region of the laser beam, this potential energy is transformed into electronic translation energy. For a Neodymium laser ($\omega \approx 1.17\text{eV}$), one has $E^2/4\omega^2 c^2 \approx 10^{-13} I \text{ eV}$, where the intensity I is given in W/cm^2 . This shows that for intensities around 10^{13} W/cm^2 this term starts to become important.

The infinite sum over Bessel functions, in Eq. (14),

can be evaluated using the integral representation

$$\sum_{n=-\infty}^{\infty} J_n(a) J_{N+2n}(b) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\theta \exp\{i(N\theta - b\sin\theta + a\sin 2\theta)\}.$$

Defining the total multiphoton transition rate by integrating Eq. (14) over the electron final momenta \vec{p} . One has

$$W^{(N)} = 2\pi \int d^3p \delta(N\omega - I_0 \left(1 + \frac{1}{2\gamma^2}\right) - \frac{p^2}{2}) |F_N(\vec{p})|^2$$

where

$$F_N(\vec{p}) = \frac{\phi_0(\vec{p})}{(2\pi)^{3/2}} \left(I_0 + \frac{p^2}{2}\right) \int_{-\pi}^{\pi} d\theta \exp\left\{i \frac{I_0}{\omega} \left[\left[1 + \frac{1}{2\gamma^2} + \frac{p^2}{2I_0}\right]\theta - \left(\frac{2}{I_0}\right)^{1/2} \frac{p \cdot \hat{x}}{\gamma} \sin\theta + \frac{1}{4\gamma^2} \sin 2\theta\right]\right\} \quad (15)$$

The integral in Eq. (15) can be evaluated by the saddle point method [7,10,11,19], under the condition $I_0/\omega \gg 1$, and in the region where $\gamma \rightarrow 0$ the amplitude of the EMF (E) must be much smaller than $9E_0$ [19]. This last condition imposes a high intensity limit on the region of applicability of Keldysh formula. Under these conditions one has

$$|F_N(\vec{p})|^2 = \frac{\gamma}{(2\pi)^4} \frac{\omega}{I_0} \frac{|\tilde{\phi}_0(\vec{p})|^2}{(1+\gamma^2)^{1/2}} \left(I_0 + \frac{p^2}{2}\right)^2 \left\{1 + (-1)^N \cos\left[\left(\frac{4I_0}{\omega}\right) \frac{(1+\gamma^2)^{1/2}}{\gamma} \times \frac{p \cdot \hat{x}}{\sqrt{2I_0}}\right]\right\} \exp\left\{-\frac{2}{3} \frac{E_0}{E} g(\gamma)\right\} \exp\left\{-\frac{2I_0}{\omega} \left(\frac{p^2}{2I_0} \operatorname{sh}^{-1} \gamma - \frac{p \cdot \hat{x}}{2I_0} \frac{\gamma}{(1+\gamma^2)^{1/2}}\right)\right\} \quad (16)$$

and

$$g(\gamma) \equiv \frac{3}{2\gamma} \left[\left(1 + \frac{1}{2\gamma^2}\right) \operatorname{sh}^{-1} \gamma - \frac{(1+\gamma^2)^{1/2}}{2\gamma}\right]. \quad (17)$$

The total transition rate is then evaluated by summing over all multiphoton processes $W_{fi} = \sum_N W_{fi}^{(N)}$, yielding a tunneling like expression for $\gamma \ll 1$.

$$W_{fi} \approx f\left(\frac{E}{E_0}\right) \exp\left[-\frac{2}{3} \frac{E_0}{E} \left(1 - \frac{\gamma^2}{10} + \dots\right)\right] \quad (18)$$

Except for the pre-exponential factor Eq.(17) reduces to the static field case for $\gamma \rightarrow 0$. The discrepancy in the pre-exponential factor may be due to the fact that the long range nature of the Coulomb force has not been properly taken into account. The specific form of $f(E/E_0)$ depends on the bound state wave function and for short range potentials Eq.(18) reproduces exactly (for $\gamma \rightarrow 0$) the adiabatic result of Sec.II [10,11].

There is still another point that must be commented which concerns the proper tunneling like exponential behavior of the ionization rate (Eq.18). The WKB result, which is recovered from Eq.(16), in the limit $\gamma \rightarrow 0$ or $\omega \rightarrow 0$, is valid for field amplitudes satisfying the condition $E \ll E_0$. This brings the question of the region of applicability of the steepest descent method, which has been used to obtain Eq.(16) and consequently the adiabatic limit of Eq.(18). It has been shown [19] that for $\gamma \rightarrow 0$ and arbitrary field intensities the infinite summation of Eq.(14) cannot be evaluated by the usual saddle point method, as applied by Keldysh [7], due to the presence of coalescing saddle points. In fact for $\gamma \rightarrow 0$, the region of applicability of Keldysh treatment is restricted to field amplitudes such that $E \ll 9E_0$ [19].

Finally we would like to discuss very briefly the

results concerning circular polarized laser light. The transition matrix (5) can be evaluated considering the same interaction Hamiltonian of Eq. (12), with the vector potential

$$\hat{A}(t) = \frac{c}{\omega} E(\hat{x} \cos \omega t + \hat{y} \sin \omega t) \quad (19)$$

The transition matrix (5) can be evaluated and for this case yields a transition rate associated to N-photon processes given by

$$W_{fi}^{(N)} = \frac{1}{2\pi} |\dot{\phi}_0(p_N^2)|^2 \left(I_0 + \frac{p_N^2}{2} \right)^2 p_N \int_0^\pi J_N^2 \left(\frac{E_0 p_N}{\omega^2} \sin \theta \right) \sin \theta d\theta \quad (20)$$

where from energy conservation

$$p_N^2 = 2 [N\omega - I_0 (1 + 1/\gamma^2)] \quad (21)$$

To obtain Eq. (20) we have assumed, for the sake of simplicity, that the initial bound state is spherically symmetrical (S-type). A general expression can also be obtained for other cases.

The integral appearing in Eq. (20) can be evaluated with the help of a table of integrals involving Bessel functions [22]

$$\int_0^\pi J_N^2(\lambda \sin \theta) \sin \theta d\theta = \frac{(\frac{\lambda}{2}) \sqrt{\pi}}{\Gamma(N+1) \Gamma(N+\frac{3}{2})} {}_1F_2 \left(\begin{matrix} N+\frac{1}{2} \\ 2N+1, N+\frac{3}{2} \end{matrix} \middle| -\lambda^2 \right) \quad (22)$$

Using the power series definition of the generalized hypergeometric function, ${}_1F_2$, [22]

$${}_1F_2 \left(\begin{matrix} N+\frac{1}{2} \\ 2N+1 \end{matrix} \middle| -\lambda^2 \right) = \frac{\Gamma(2N+1) \Gamma(N+3/2)}{\Gamma(N+1/2)} \sum_{k=0}^{\infty} (-1)^k \frac{\Gamma(N+1/2+k) \lambda^{2k}}{\Gamma(2N+1+k) \Gamma(N+\frac{3}{2}+k) k} \quad (23)$$

it is easily shown that this function can be written in terms of an integral over a Bessel function, i.e.

$$\int_0^\pi J_N^2(\lambda \sin\theta) \sin\theta d\theta = 2\lambda^{-1} \int_0^\lambda J_{2N}(2\delta) d\delta. \quad (24)$$

This last equation shows that, except for the choice of gauge, this result is equivalent to the one obtained by Perelomov et al [10]. The integral on the right hand side of Eq. (24) may be evaluated in a similar manner as done in [10]. If one introduces the variables,

$$v_c \equiv (I_0/\omega)(1 + 1/\gamma^2) \quad \text{and} \quad t \equiv (2v_c/N) - 1,$$

Eq. (20) can be rewritten as

$$W_{fi}^{(N)} = \frac{\sqrt{2I_0}}{\pi} |\tilde{\phi}_0(p_N^2)|^2 \left(I_0 + \frac{p_N^2}{2}\right)^2 \frac{1+\gamma^2}{\gamma(1+t)} \int_0^\lambda \left[\frac{(1-t^2)}{(1+\gamma^2)}\right]^{\frac{1}{2}} J_{2N}(2N\delta) d\delta \quad (25)$$

For $I_0 \gg \omega$ we have $N \gg 1$, and the Bessel function of Eq. (25) has large values of the argument and of the index. An asymptotic expression for this case can be used [22] and Eq. (25) can be transformed [10] into

$$W^{(N)} = \frac{\sqrt{2I_0}}{4\pi^{\frac{3}{2}} (2v_c)^{\frac{3}{2}}} |\tilde{\phi}_0(p_N^2)|^2 \left(I_0 + \frac{p_N^2}{2}\right)^2 (1+t) \left[\left(1 + \frac{1}{\gamma^2}\right) (1-t)\right]^{\frac{1}{2}} \left(\frac{1+\gamma^2}{\gamma^2 + t^2}\right)^{\frac{3}{4}} \times \exp\{-4v_c \phi(t, \gamma)\} \quad (26)$$

where [10]

$$\phi(t, \gamma) = \frac{1}{1+t} \left\{ \text{th}^{-1} \left(\frac{t^2 + \gamma^2}{1 + \gamma^2} \right) - \left(\frac{t^2 + \gamma^2}{1 + \gamma^2} \right) \right\} \quad (27)$$

Following Perelomov et al [10], it is easy to show that the total transition rate can be written in the form

$$W = \sum_N W^{(N)} = f\left(\frac{E_0}{E}, \gamma\right) \exp\left\{-\frac{2}{3} \frac{E_0}{E} g_c(\gamma)\right\} \quad (28)$$

where for $\gamma \rightarrow 0$, $g_c(\gamma) \rightarrow 1 - \gamma^2/15$, reproducing also a tunnelling like behavior. For a ruby laser, linearly polarized light should ionize atoms more strongly than circularly light [10], but for CO₂ or Neodimium lasers the probabilities are basically the same.

In conclusion we must mention that the type of non-perturbative approach presented here seems to be convenient to describing several different regions of approximations, including the strong field limit. The experimental results of Boreham et al [6] and Baldwin and Boreham [8], performed in the region $0.1 \leq \gamma \leq 1$ and $E = E_0$, shows deviations from the predictions of perturbation theory and at the moment tends to support the Keldysh type of approach discussed in this work.

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