Theor Chem Acc (2010) 127:175–182
DOI 10.1007/s00214-009-0712-z

REGULAR ARTICLE

S-matrix theory of high harmonic generation and ionization of coherently ro-vibrating linear molecules by intense ultrashort laser pulses

F. H. M. Faisal

Received: 16 August 2009 / Accepted: 30 November 2009 / Published online: 22 December 2009

Abstract A theory of high harmonic generation and ionization of coherently rotating and vibrating linear molecules by a delayed pair of intense ultrashort laser pulses is presented. It correlates the nuclear motions in real time with the modulation of the harmonic emission and ionization signals as a function of the time-delay between the exciting and the probing pulses. An illustrative analytical example of the excitation and detection of the clock-motion associated with the "0–1" vibrational oscillation of a diatomic molecule is also given.

Keywords S-Matrix theory · High harmonic generation, · Ionization · Ro-vibration · Intense · Ultrashort · Laser pulses · Pump-prove signal · Time-delay · Clock-motion

1 Introduction

High harmonic generation (HHG) and ionization are two most conspicuous phenomena associated with interaction of molecules with very intense ultrashort (femtosecond to attosecond) laser pulses. In the dynamic HHG experiments (e.g., [1–6]), coherent nuclear motions are induced and monitored by two intense ultrashort laser pulses, one of which (the "pump" pulse) sets the molecules in coherent rotational motion, and a second pulse (the "probe" pulse) causes the rotating molecule to emit high harmonic radiations (at a multiple of the laser frequency). The HHG emission intensity is recorded as a function of the time-delay between the pump and the probe pulses, and thus it serves as the signal that monitors the underlying coherent nuclear motion. Current theoretical interests in molecular HHG are increasingly moving to explore (e.g., [7–12]) the ultrafast time-dependent effects of nuclear motions on the HHG signals. Recently, a quantum S-matrix theory [8–10] of intense-field pump–probe experiments and analysis of the dynamic HHG signals observed in the experiments have been given. The S-matrix theory is shown to reproduce all the salient characteristics of rotational coherence, dynamic alignment, and HHG signals observed both in time and frequency domains. They also reproduce the dependence of the dynamic HHG signal on the relative angle between the polarization axes of the pump and the probe pulses. The theory has also predicted the existence of a symmetry-dependent "magic angle" ($\arctan\sqrt{2} \approx 55^\circ$) for the relative polarization directions, for which the dynamic signal becomes independent of the time-delay and produces a uniform HHG signal. This effect being generic in nature can help to identify the active molecular orbital symmetry of an unknown linear molecule [9]. Both the existence of the "magic angle" and its neighborhood and their *generic* dependence on the orbital symmetry, as predicted, have now been seen experimentally [5, 6]. In view of the fruitfulness of the intense-field S-matrix theory, in this paper, we extend the theory to incorporate the vibrational motion, and derive a mathematical expression for the dynamic HHG signal for future analyses of simultaneous ro-vibrational coherence in intense-field pump–probe experiments. Before concluding, we also give an explicit example of vibrational coherence (in the form of a "clock-motion") that can be induced and detected by observing the modulation of the *ionization* signal from a diatomic molecule.

Dedicated to the memory of Professor Jürgen Hinze and published as part of the Hinze Memorial Issue.

F. H. M. Faisal (✉)
Fakultät für Physik, Universität Bielefeld,
Postfach 100131, 33501 Bielefeld, Germany
e-mail: ffaisal@physik.uni-bielefeld.de

2 Theoretical formulation

Both HHG and ionization of atoms and molecules by strong laser fields can be described fruitfully within the intense-field many body S-matrix theory (or IMST), see [13] for a review. IMST provides a means of systematically rearranging the usual S-matrix series in such a way that the most relevant highly non-perturbative contributions of the laser interaction in a given process of interest may appear in the leading terms of the new series. For example, for the ionization process, the leading term of the IMST series corresponds to the so-called strong-field KFR1 approximation that provides, in the limiting case of high intensity and/or low frequency, the well-known "optical tunneling" mechanism of ionization of atoms and molecules in intense laser fields. Its application to molecules led to the discovery of the symmetry-dependent molecular two-slit interference effect and the related phenomenon of "suppressed ionization" of molecules [14] in strong laser fields. The second leading term of IMST (or KFR2 approximation) yielded the first quantum mechanical theory of non-sequential double ionization (NSDI) [15, 16] and of high harmonic generation [17]. They also provided the quantum mechanical justifications and basis for the intuitively appealing classical "re-scattering" mechanism [18, 19].

The fundamental quantity in quantum electrodynamics (QED) of emission of harmonic radiation is the probability amplitude for the photon emission process [20, 21]. It has been demonstrated explicitly within the intense-field S-matrix theory recently (e.g., [10]) that the Fourier transform (FT) of the time-dependent expectation value of the electronic dipole operator is *proportional* to the quantum transition amplitude for the HHG process of present interest. Therefore, toward the goal of deriving a mathematical expression for the dynamic HHG signal exhibiting the ro-vibrational coherence in mind, we shall first derive the time dependent expectation value of the electronic dipole operator.[1] We shall then construct the dynamic HHG signals from coherently ro-vibrating linear molecules. Finally, in Sect. 4 we shall consider the dynamic ionization signals.

3 Molecular HHG with time-delayed interactions

The total Hamiltonian of the system consisting of a molecule interacting with a pump pulse L_1 at a time t and a probe pulse L_2 at a delayed time $t - t_{\mathrm{d}}$, in the adiabatic Born–Oppenheimer approximation, can be written (in a.u.: $e = \hbar = m = \alpha c = 1$) as [8–10]:

$$H_{\mathrm{tot.}}(t) = H_N^{(0)} + V_{N-L_1}(t) + H_e^{(0)} + V_{e-L_2}(t - t_{\mathrm{d}}) \tag{1}$$

where $H_N^{(0)}$ is the nuclear Hamiltonian, $V_{N-L_1}(t)$ the interaction due to the pump-pulse L_1 with the nuclear motion, $H_e^{(0)}$ is the electronic Hamiltonian, and $V_{e-L_2}(t - t_{\mathrm{d}})$ is the delayed interaction of the probe-pulse L_2 with the active electron. We recall that in this approximation the electronic wavefunctions depend adiabatically on the nuclear coordinates, $\boldsymbol{R}$, such that their gradients, or derivatives with respect to $\boldsymbol{R}$, and hence the non-adiabatic couplings, are assumed to be zero. We describe the laser fields in terms of the vector potentials which have the form $\dot{\boldsymbol{A}}(t) = -c\boldsymbol{F}(t)$, where $\boldsymbol{F}(t)$ is the electric field of the laser at time t. Thus, the interaction Hamiltonians appearing above (assuming the usual dipole approximation for the laser fields, and negligible overlap between the pump and probe pulses) are

$$V_{N-L_1}(t) = -\boldsymbol{\mu} \cdot \boldsymbol{F}^{(1)}(t) - \frac{1}{2}\sum_{ij} F_i^{(1)}(t)\alpha_{ij}F_j^{(1)}(t) \tag{2}$$

and

$$V_{e-L_2}(t - t_{\mathrm{d}}) = -\hat{\boldsymbol{d}}_e \cdot \boldsymbol{F}^{(2)}(t - t_{\mathrm{d}}), \tag{3}$$

where $\boldsymbol{\mu}$ is the permanent dipole moment (if non-zero), and α_{ij} is the polarization *tensor* of the molecule, and $\hat{\boldsymbol{d}}_e$ is the electronic transition dipole operator; $\boldsymbol{F}^{(1)}(t)$ and $\boldsymbol{F}^{(2)}(t)$ are the pump and the probe pulses, respectively.

The Schrödinger equation of the total interacting system is

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = H_{\mathrm{tot.}}(t)|\Psi(t)\rangle \tag{4}$$

and the total Green's function of the system, $G(t, t')$, is defined by

$$\left(i\frac{\partial}{\partial t} - H_{\mathrm{tot.}}(t)\right)G(t, t') = \delta(t - t')\mathbf{1}. \tag{5}$$

To obtain the desired solution of Eq. 4, we first construct the total Green's function by considering both the nuclear and the electronic motions in the laser fields.

---

[1] QED ensures that the quantum minimal coupling prescription (and/or any of its gauge equivalent form) can be used consistently to treat high harmonic generation from molecules. In particular, this is shown explicitly for the S-matrix theory, either in the so-called "velocity gauge" [13] and/or in the "length gauge" [10] that are connected to each other by a gauge transformation. Moreover, the theoretical results obtained recently from the quantum length gauge formulation within the S-matrix theory [10] are in excellent agreement with recent experimental results for dynamic HHG signals from linear molecules (e.g., [6, 10]). Thus, there is no essential need to use the so-called "acceleration" expectation value. This is moreover not connected to the quantum minimal-coupling prescription by a gauge transformation (but it is often motivated by invoking Ehrenfest's theorem, or by the well-known Larmor expression for the power emitted according to classical electrodynamics which is known of course to be not fully satisfactory). In some theoretical formulations, "acceleration" expectation values have been used apparently to avoid certain technical difficulties (e.g., [12] and references cited therein).

3.1 Driven nuclear motion

The driven nuclear motion can be described in terms of the complete set of *fundamental* (linearly independent) solutions of the Schrödinger equation,

$$i\frac{\partial}{\partial t}|\Phi_{vJM}(t)\rangle = (H_N^{(0)} + V_{N-L_1}(t))|\Phi_{vJM}(t)\rangle. \tag{6}$$

They are obtained by solving the coupled time-dependent differential equations satisfied by the expansion coefficients $C_{vJM}^{v_0j_0M_0}(t)$ of $|\Phi_{v_0J_0M_0}(t)\rangle$, where

$$|\Phi_{v_0J_0M_0}(t)\rangle = \sum_{vJM} C_{vJM}^{v_0J_0M_0}(t)\mathrm{e}^{-iE_{vJM}t}|vJM\rangle. \tag{7}$$

For the sake of simplicity and to be concrete, we have restricted ourselves above to the basis functions appropriate for a linear molecule, namely the angular momentum eigenfunctions, $|JM\rangle$, and, say, the Morse vibrational wavefunctions, $|v\rangle$ (clearly, of course, for a more general molecular geometry, they should be replaced correspondingly by the appropriate nuclear basis functions). The linear differential equations of interest are

$$\begin{aligned} &i\frac{\partial}{\partial t}C_{vJM}^{v_0J_0M_0}(t) \\ &\quad = \sum_{v'J'M'} \langle vJM|V_{N-L_1}(t)|v'J'M'\rangle \mathrm{e}^{i(E_{vJM}-E_{v'J'M'})t} C_{v'J'M'}^{v_0J_0M_0}(t). \end{aligned} \tag{8}$$

The set of fundamental solutions $|\Phi_{v_0J_0M_0}(t)\rangle$ is obtained from the initial condition $|\Phi_{v_0J_0M_0}(0)\rangle = |v_0J_0M_0\rangle$, i.e., from the solutions of Eq. 8 with the initial conditions $C_{vJM}^{v_0J_0M_0}(t=0) = \delta_{v,v_0}\delta_{J,J_0}\delta_{M,M_0}$. Equation 7 represents a ro-vibrational wave-packet state that evolves in one-to-one correspondence with the initial, e.g., thermally occupied ro-vibrational eigenstates of the unperturbed molecule. It should be recalled that these wave-packet states are linearly independent, and like the eigenstates $|vJM\rangle$ of $H_N^{(0)}$, they form a complete set:

$$\sum_{vJM} |\Phi_{vJM}(t)\rangle\langle\Phi_{vJM}(t)| = \mathbf{1}. \tag{9}$$

3.2 Driven electronic motion

The driven electronic motion arising from the interaction of the active electron with an intense probe pulse L_2 is governed by the electronic Green's function (cf. [9, 10]) $G_e(t, t')$, which satisfies the equation,

$$\left\{i\frac{\partial}{\partial t} - (H_e^{(0)} + V_{e-L_2}(t-t_\mathrm{d}))\right\}G_e(t,t') = \delta(t-t')\mathbf{1}. \tag{10}$$

We solve the above equation using the molecular KFR approximation as (cf. [10]):

$$\begin{aligned} G_e(t,t') &= -i\theta(t-t')\sum_{j,\boldsymbol{p}} |\phi_j^{(+)}(t)\rangle|\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t-t_\mathrm{d}))\rangle \\ &\quad \times \langle\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t'-t_\mathrm{d}))|\langle\phi_j^{(+)}(t')| \\ &= -i\theta(t-t')\sum_{j,\boldsymbol{p}} |\phi_j^{(+)}(t)\rangle|\tilde{\boldsymbol{p}}(t-t_\mathrm{d})\rangle \mathrm{e}^{-\frac{i}{2}\int_{t'-t_\mathrm{d}}^{t-t_\mathrm{d}} \boldsymbol{p}^2(u)\mathrm{d}u} \\ &\quad \times \langle\tilde{\boldsymbol{p}}(t'-t_\mathrm{d})|\langle\phi_j^{(+)}(t')|, \end{aligned} \tag{11}$$

where the index j runs over the electronic states of the molecular ion, and we have defined the "asymptotic Coulomb–Volkov" states $|\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}, t)\rangle$ by [22]:

$$|\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t))\rangle = |\tilde{\boldsymbol{p}}(t)\rangle \mathrm{e}^{-\frac{i}{2}\int^{t} \boldsymbol{p}^2(u)\mathrm{d}u}, \tag{12}$$

where in coordinate representation,

$$\langle \boldsymbol{r}|\tilde{\boldsymbol{p}}(t)\rangle = \mathrm{e}^{i\boldsymbol{p}(t)\boldsymbol{r}+i\eta(t)\ln(2p(t)r)}. \tag{13}$$

$\boldsymbol{p}$ is the free momentum of the electron, and $\boldsymbol{p}(t)$ stands for the instantaneous electron momentum in the presence of the field: $\boldsymbol{p}(t) \equiv (\boldsymbol{p} + \boldsymbol{A}(t)/c); \eta(t) = \frac{Z}{p(t)}$ is a time-dependent Sommerfeld-parameter and Z is the asymptotic nuclear charge. We note that the approximate "asymptotic Coulomb–Volkov" wavefunction of the active electron, which goes over to the usual Volkov wavefunction for $Z = 0$, form effectively a complete set of states (i.e., to within the approximation that the slowly varying logarithmic phase factor is assumed to change negligibly during the completeness integration):

$$\sum_{\boldsymbol{p}} |\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t))\rangle\langle\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t))| = \mathbf{1}. \tag{14}$$

3.3 Total Green's function

Thus, using Eqs. 9 and 11, we write down the total Green's function $G(t, t')$ associated with the Hamiltonian $H_{\mathrm{tot.}}(t)$ as,

$$\begin{aligned} G(t,t') &= -i\theta(t-t') \\ &\quad \times \sum_{vJM,j,\boldsymbol{p}} |\phi_j^{(+)}\rangle|\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t-t_\mathrm{d}))\rangle|\Phi_{vJM}(t)\rangle \mathrm{e}^{-iE_j^{+}(t-t')} \\ &\quad \times \langle\Phi_{vJM}(t')|\langle\phi_{cv}^{(-)}(\boldsymbol{p}(t'-t_\mathrm{d}))|\langle\phi_j^{(+)}|, \end{aligned} \tag{15}$$

where $|\phi_{\mathrm{cv}}^{(-)}(\boldsymbol{p}(t))\rangle$ are the asymptotic Coulomb–Volkov states defined above, and $|\phi_j^{(+)}\rangle$ stands for the molecular orbitals of the residual ion. The validity of the above construction of the Green's function (15) can be readily established by substituting it in Eq. 5 and (consistently with the B-O adiabatic approximation) by neglecting the derivatives of the electronic orbitals with respect to the nuclear coordinates $\boldsymbol{R}$.

3.4 The state of the total system

We require the state of the total system that satisfies the initial condition according to which the nuclear motion is prepared (by the pump laser L_1) in a ro-vibrational wavepacket state $|\Phi_{V_0 J_0 M_0}(t)\rangle$, while the electron motion is in the active molecular orbital(s) of the ground electronic state $|\phi_0(1, 2, ..., N_e;t)\rangle$. Thus, the initial state of the total system is

$$|\Psi_0(t)\rangle = |\phi_0(1,2,...,N_e;t)\rangle|\Phi_{v_0 J_0 M_0}(t)\rangle, \tag{16}$$

where N_e is the number of equivalent electrons in the ground state. Using Eq. 15, we can write the state of the system that evolves from the initial state Eq. 16 (after the interaction with the probe laser L_2) as

$$|\Psi(t)\rangle = |\Psi_0(t)\rangle + \int^{t} \mathrm{d}t' G(t,t') V_{e-L_2}(t'-t_\mathrm{d})|\Psi_0(t')\rangle. \tag{17}$$

3.5 Dipole expectation value and HHG transition amplitude

The quantum probability amplitude for the emission of a high harmonic photon, as indicated earlier, is proportional to the FT of the expectation value of the electronic dipole operator [10]. The expectation value, $\boldsymbol{D}(t)$, of the dipole operator associated with the coupling of the probe laser L_2 with the active electron of the molecule, $\hat{\boldsymbol{d}}_e$, is

$$\begin{aligned}\boldsymbol{D}(t) &= \langle\Psi(t)|\hat{\boldsymbol{d}}_e|\Psi(t)\rangle \\ &= \langle\Psi_0(t)|\hat{\boldsymbol{d}}_e|\Psi_0(t)\rangle + \Bigg\{\langle\Psi_0(t)|\hat{\boldsymbol{d}}_e \int_0^t \mathrm{d}t' G(t,t') \\ &\quad \times V_{e-L_2}(t'-t_\mathrm{d})|\Psi_0(t')\rangle + \text{c.c.}\Bigg\} + \cdots \\ &= \langle\Psi_0(t)|\hat{\boldsymbol{d}}_e \int_0^t \mathrm{d}t' G(t,t') V_{e-L_2}(t'-t_\mathrm{d})|\Psi_0(t')\rangle + \text{c.c.},\end{aligned} \tag{18}$$

where c.c. stands for the complex conjugate of the term preceding it. In the last line, we have dropped the first term (it corresponds to a static term or vanishes for a fixed parity initial state) and neglected the quadratic term (consistent with the lowest order KFR approximation [10]). Substituting Eq. 15 in 18 we get more explicitly,

$$\begin{aligned}\boldsymbol{D}(t) = -i \sum_{vJM,j} &\langle\Psi_{v_0 J_0 M_0}(t) \\ &\times |\langle\phi_j^{(+)}(1,2,..,N_e-1;t)|\hat{\boldsymbol{d}}_e|\Phi_{vJM}(t)\rangle \int_0^t \mathrm{d}t' G_e(t,t') \\ &\times \langle\Phi_{vJM}(t')|V_{e-L_2}(t'-t_\mathrm{d})|\phi_0(1,2,..,N_e;t')\rangle| \\ &\times \Phi_{v_0 J_0 M_0}(t')\rangle + \text{c.c.}\end{aligned} \tag{19}$$

In the above, we have neglected the interaction of the *tightly* bound ionic electrons with the probe laser field L_2 ("bare ion" approximation). If and when required, it could be included by replacing the bare ionic states by the corresponding "dressed" ionic states. The so-called Dyson orbitals are defined by the overlap of the initial molecular ground state and the ionic states, or

$$\begin{aligned}|\phi_j^{(D)}(t)\rangle &= \langle\phi_j^{(+)}(1,2,...,N_e-1;t)|\phi_0(1,2,...,N_e;t)\rangle \\ &= \langle\phi_j^{(+)}(1,2,...,N_e-1)|\phi_0(1,2,...,N_e)\rangle \mathrm{e}^{-i\epsilon_j t} \\ &\equiv |\phi_j^{(D)}\rangle \mathrm{e}^{-i\epsilon_j t}\end{aligned} \tag{20}$$

where $\epsilon_j \equiv |E^{(+)}{}_j - E^{(0)}|, j = 0, 1, 2, ...$ are the successive ionization potentials of the molecule. In terms of the Dyson orbitals, we may simplify the expectation value

$$\begin{aligned}\boldsymbol{D}(t) = -i \sum_{vJM,j,\boldsymbol{p}} &\langle\Phi_{v_0 J_0 M_0}(t)|\langle\phi_j^{(D)}(t)|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t-t_\mathrm{d})\rangle|\Phi_{vJM}(t)\rangle \\ &\times \mathrm{e}^{-\frac{i}{2}\int^{t-t_\mathrm{d}} \boldsymbol{p}^2(u)\mathrm{d}u} \int_0^t \mathrm{d}t' \mathrm{e}^{\frac{i}{2}\int^{t'-t_\mathrm{d}} \boldsymbol{p}^2(u)\mathrm{d}u} \\ &\times \langle\tilde{\boldsymbol{p}}(t'-t_\mathrm{d})|\langle\Phi_{vJM}(t')|V_{e-L_2}(t'-t_\mathrm{d}) \\ &\times |\phi_j^{(D)}(t')\rangle|\Phi_{v_0 J_0 M_0}(t')\rangle + \text{c.c.}\end{aligned} \tag{21}$$

We may add parenthetically that the Dyson orbitals, $|\phi_j^{(D)}\rangle$, and the corresponding energies, ϵ_j, implicitly include effects of electron–electron correlation that the usual Hartree–Fock orbitals cannot. The present theory incorporates the Dyson orbitals in a natural way. Clearly, therefore, they can be used if desired to replace the more commonly used Hartree–Fock MOs and the associated HOMO, HOMO-1,..., etc., in the present context. We have found it also useful (e.g., in the related context of the "inverse problem" of molecular orbital retrieval (e.g., [7]) to refer to the sequence of Dyson orbitals $|\phi_j^{(D)}\rangle$, $j = 0, 1, 2, ...$, as HODO, HODO-1,..., etc., to distinguish them from the corresponding Hartree–Fock orbitals. The dipole expectation value now takes the form

$$\begin{aligned}\boldsymbol{D}(t) = -i \sum_{vJM,j,\boldsymbol{p}} &\langle\Phi_{v_0 J_0 M_0}(t)|\langle\phi_j^{(D)}|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t-t_\mathrm{d})\rangle|\Phi_{vJM}(t)\rangle \\ &\times \int_0^t \mathrm{d}t' \mathrm{e}^{-i\int_{t'-t_\mathrm{d}}^{t-t_\mathrm{d}}(\boldsymbol{p}^2(u)/2+\epsilon_j)\mathrm{d}u} \langle\tilde{\boldsymbol{p}}(t'-t_\mathrm{d}) \\ &\times |\langle\Phi_{vJM}(t')|V_{e-L_2}(t'-t_\mathrm{d}) \\ &\times |\phi_j^{(D)}\rangle|\Phi_{v_0 J_0 M_0}(t')\rangle + \text{c.c.}\end{aligned} \tag{22}$$

We change the variable, $t' \to t_\mathrm{d} + t'$, and similarly, $t \to t_\mathrm{d} + t$, where t_d is the time-delay between the pump and the probe pulses L_2 (it is usually defined by the difference between the peaks of the pump and the probe pulses). We may thus rewrite the above expression as

$$\boldsymbol{D}(t) = -\,i \sum_{vJM,j,\boldsymbol{p}} \langle \Phi_{v_0J_0M_0}(t_\mathrm{d})|\mathrm{e}^{iH_N^{(0)}t}\langle \phi_j^{(D)}|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t)\rangle \mathrm{e}^{-iH_N^{(0)}t}|\Phi_{vJM}(t_\mathrm{d})\rangle$$
$$\times \int_{t_\mathrm{d}}^{t_\mathrm{d}+t} \mathrm{d}t' \mathrm{e}^{-i\int_{t'}^{t}(\boldsymbol{p}^2(u)/2+\epsilon_j)\mathrm{d}u} \langle \Phi_{vJM}(t_\mathrm{d})|\mathrm{e}^{iH_N^{(0)}t'}$$
$$\times \langle \tilde{\boldsymbol{p}}(t')|V_{e-L_2}(t')|\phi_j^{(D)}\rangle \mathrm{e}^{-iH_N^{(0)}t'}|\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle + \mathrm{c.c.} \tag{23}$$

where $H_N^{(0)}$ is the nuclear (ro-vibrational) Hamiltonian. Assuming that the highest excitation energy of the ro-vibrational spectrum during the process of interest lies far below the electronic excitation energy, i.e., for

$$\mathrm{Max}(\Delta E_{vJM,v'J'M'}) \ll \epsilon_{j=0}$$
$$\equiv I_p(\text{the first ionization potential}), \tag{24}$$

which is well satisfied in practice, the emission of a given harmonic becomes a quasi-elastic process with respect to the ro-vibrational transitions within the nuclear wave-packet states. Neglecting the non-adiabatic couplings between the nuclear and the electronic coordinates (within the present Born–Oppenheimer approximation) we may commute the nuclear Hamiltonian $H_N^{(0)}$ with the electronic states (and the electronic dipole operators) and further simplify the expectation value as:

$$\boldsymbol{D}(t) = -i \sum_{vJM,j,\boldsymbol{p}} \langle \Phi_{v_0J_0M_0}(t_\mathrm{d})|\langle \phi_j^{(D)}|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t)\rangle$$
$$\times \int_{t_\mathrm{d}}^{t_\mathrm{d}+t} \mathrm{d}t' \mathrm{e}^{-i\int_{t'}^{t}(\boldsymbol{p}^2(u)/2+\epsilon_j)\mathrm{d}u}$$
$$\times |\Phi_{vJM}(t_\mathrm{d})\rangle\langle \tilde{\boldsymbol{p}}(t')|\langle \Phi_{vJM}(t_\mathrm{d})|V_{e-L_2}(t')|\phi_j^{(D)}\rangle$$
$$\times |\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle + \mathrm{c.c.} \tag{25}$$

In view of the completeness relation, Eq. 9, the sum over the final wave-packet states can be carried out immediately and we get,

$$\boldsymbol{D}(t) = -i\langle \Phi_{v_0J_0M_0}(t_\mathrm{d})|D_e(t,\boldsymbol{R})|\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle \tag{26}$$

where we have defined the time-dependent electronic part of the dipole expectation value by

$$\boldsymbol{D}_e(t,\boldsymbol{R}) = \sum_{j,\boldsymbol{p}} \langle \phi_j^{(D)}|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t)\rangle$$
$$\times \int_{t_\mathrm{d}}^{t_\mathrm{d}+t} \mathrm{d}t' \mathrm{e}^{-i\int_{t'}^{t}(\boldsymbol{p}^2(u)/2+\epsilon_j)\mathrm{d}u}$$
$$\times \langle \tilde{\boldsymbol{p}}(t')|V_{e-L_2}(t')|\phi_j^{(D)}\rangle + \mathrm{c.c.} \tag{27}$$

We may note that in the drastic "frozen nuclei" approximation, one would take the $\boldsymbol{R}$-(nuclear separation and orientation) dependent integrand in Eq. 26 outside the integration with respect to the ro-vibrational wavepacket states. However, in the less drastic "adiabatic nuclei" approximation [23, 24], only the *gradients* of the nuclear coordinate dependence are neglected but the integrations over the nuclear coordinates as such are performed as they appear in Eq. 26. We may point out in this context that recent observation of the so-called "anomalous" rotational series and lines in the Fourier spectra of dynamic high harmonic signals and their analysis have confirmed [8] the ability of the "adiabatic nuclei" approximation to account for their presence, while the corresponding calculations using the "frozen nuclei" approximation could not account for them (see sec. VII D [10]). Next, we take the FT of $\boldsymbol{D}_e(t, \boldsymbol{R})$ with respect to the frequency, Ω, and write

$$\boldsymbol{D}_e(t,\boldsymbol{R}) = -2\pi i \int \mathrm{d}\Omega \mathrm{e}^{-i\Omega t}\tilde{\boldsymbol{D}}_e(\Omega,\boldsymbol{R}) + \mathrm{c.c.} \tag{28}$$

We define $\tilde{\boldsymbol{D}}_e(\Omega = n\omega, \boldsymbol{R}) \equiv \tilde{\boldsymbol{D}}_e^{(n)}(\boldsymbol{R})$, which is the value of the FT at the nth peak, $\Omega = n\omega$, where ω is the fundamental laser frequency. Or,

$$\tilde{\boldsymbol{D}}_e^{(n)}(\boldsymbol{R}) = \mathrm{FT}\left\{ \sum_{j,\boldsymbol{p}} \langle \phi_j^{(D)}|\hat{\boldsymbol{d}}_e|\tilde{\boldsymbol{p}}(t)\rangle \right.$$
$$\left. \times \int_{t_\mathrm{d}}^{t_\mathrm{d}+t} \mathrm{d}t' \mathrm{e}^{-i\int_{t'}^{t}(\boldsymbol{p}^2(u)/2+\epsilon_j)\mathrm{d}u} \langle \tilde{\boldsymbol{p}}(t')|V_{e-L_2}(t')|\phi_j^{(D)}\rangle \right\}$$
$$\times (\Omega = n\omega) + \mathrm{c.c.} \tag{29}$$

Using the above equation we can write the FT of the expectation value of interest, as

$$\tilde{\boldsymbol{D}}_{i,i}^{(n)}(t_\mathrm{d}) = \langle \Phi_{v_0J_0M_0}(t_\mathrm{d})|\tilde{\boldsymbol{D}}_e^{(n)}(\boldsymbol{R})|\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle$$
$$\equiv \langle i, t_\mathrm{d}|\tilde{\boldsymbol{D}}_e^{(n)}(\boldsymbol{R})|i, t_\mathrm{d}\rangle, \tag{30}$$

where for the sake of brevity, we have written the ro-vibrational wavepacket states $|\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle$ as

$$|v_0J_0M_0(t_\mathrm{d})\rangle = |i, t_\mathrm{d}\rangle, \quad \text{where } i \equiv (v_0, J_0, M_0). \tag{31}$$

3.6 The dynamic HHG signal

The fundamental transition matrix element $T^{(n)}{}_{i,i}(t_\mathrm{d})$ for the emission of the nth harmonic, as mentioned earlier, is proportional to the the nth FT of the dipole expectation value, and it is given fully by (cf. [10]):

$$T_{i,i}^{(n)}(t_\mathrm{d}) = \sqrt{2\pi(n\omega)}\epsilon_\Omega \cdot \tilde{\boldsymbol{D}}_{i,i}^{(n)}(t_\mathrm{d}), \tag{32}$$

where ϵ_Ω is the unit polarization vector of the emitted harmonic radiation. Thus, the probability of emission of the nth harmonic radiation per unit time (or the *rate* of emission) from each of the ro-vibrational wavepacket states $|i, t_d\rangle = |\Phi_{v_0J_0M_0}(t_\mathrm{d})\rangle$, is readily obtained from the generalized Fermi golden rule (e.g., [21]):

$$d\Gamma_i^{(n)}(t_\mathrm{d}) = 2\pi |T_{i,i}^{(n)}(t_d)|^2 \delta(\Omega - n\omega) \frac{d^3 K_\Omega}{(2\pi)^3} \tag{33}$$

where $K_\Omega = \frac{\Omega}{c} = \frac{n\omega}{c}$ is the wave number of the nth harmonic radiation. The dynamic HHG signal for the emission of harmonic radiation from the ensemble of target molecules in the pump–probe experiments corresponds to the statistical average of the nth harmonic emission probability with respect to the initial thermal distribution of the ro-vibrational eigenstates at a gas temperature T. Thus, the dynamic HHG signal as a function of t_d is given by,

$$\frac{dS^{(n)}(t_\mathrm{d}; \hat{\boldsymbol{K}}_\Omega \parallel \hat{\boldsymbol{\kappa}}_\omega)}{d\hat{K}_\Omega} = \sum_i \rho_i |\epsilon_\Omega \cdot \tilde{\boldsymbol{D}}_{i,i}^{(n)}(t_\mathrm{d})|^2 \frac{(n\omega)^3}{2\pi c^3}, \tag{34}$$

where ρ_i is the Boltzmann distribution of the initial ro-vibrational states $i \equiv (v_0, J_0, M_0), \rho_i = \rho_{v_0 J_0 M_0} = e^{-E_{v_0 J_0 M_0}/k_B T}/Z_P$, where k_B is the Boltzmann constant, $Z_P = \sum_{v_0 J_0 M_0} e^{-E_{v_0 J_0 M_0}/k_B T}$ is the partition function (one may note that the zero-point energy corresponds to the lowest allowed eigenvalue $E_{v_0} = v_\mathrm{min}$, $J_0 = J_\mathrm{min}$, $M_0 = M_\mathrm{min}$ of the molecule). Equation 34 is the desired final expression for the dynamic HHG signal for the simultaneous excitation and detection of ro-vibrational coherence. It should be noted that if one formally drops the vibrational degrees of freedom in this result, then it reduces as required to the dynamic HHG signal for the rotational coherence derived (and investigated in great details) by us earlier [10].

Before concluding this work we briefly outline the derivation of the corresponding signal for ionization in intense-field pump–probe experiments, and provide an illustrative analytic example of vibrational coherence in the form of a "clock-motion" in a diatomic molecule, as mentioned earlier.

4 Dynamic ionization signal

Within the S-matrix theory, the pump–probe signal showing ro-vibrational coherence in the *ionization* channel can be derived in a similar way. In fact, the result can be written requiring only the ionization transition into the Volkov state $|\phi_\mathrm{cv}^{(-)}(\boldsymbol{p}(t))\rangle$. Formally, this gives an expression analogous to the first part of the generalized dipole operator, derived above, involving only the "bound-free" transition to the Volkov state. We assume that the active MO in coordinate representation can be expressed as:

$$\langle \boldsymbol{r} | \phi_0 \rangle = \sum_{i=1}^{N} \sum_{j=1}^{j_\mathrm{max}} c_j^{(i)} \phi_j^{(i)}(\boldsymbol{r} - \boldsymbol{R}_i), \tag{35}$$

where $\boldsymbol{R} \equiv \{\boldsymbol{R}_1, \boldsymbol{R}_2, ..., \boldsymbol{R}_N\}$ and $\boldsymbol{r}$ stand for the coordinates of the nuclei and the active electron, respectively, and the coefficients $c_j^{(i)}$ arise from the linear superposition of the atomic orbitals $|\phi_j^{(i)}\rangle, j = 1, 2, ..., j_\mathrm{max}$. For the sake of simplicity, one may obtain the MOs from Hartree–Fock variational approximation [25]. Then, in the lowest order (KFR1 approximation), the T-matrix for ionization by absorption of n photons from an intense field, as shown first in [14], takes the form:

$$\begin{aligned} T_\mathrm{ion}^{(n)}(\boldsymbol{p}, \boldsymbol{R}) = \frac{\omega}{2\pi} \int_0^{2\pi/\omega} & dt e^{i \int^t (\boldsymbol{p}^2(t')/2 + I_p) dt'} \\ & \times \sum_{i,j} c_j^{(i)} \int d^3 r e^{-i\eta(t) \ln(2p(t)r)} e^{-i\boldsymbol{p}(t)\cdot \boldsymbol{r}} \\ & \times (-\boldsymbol{F}(t) \cdot \boldsymbol{r}) \phi_j^{(i)}(\boldsymbol{r} - \boldsymbol{R}_i), \end{aligned} \tag{36}$$

where ω is the central frequency of the laser, $\eta(t) = \frac{Z}{\boldsymbol{p}(t)}$, and I_p is the (first) ionization potential. Using the generalized Fermi golden rule, and the transition matrix Eq. 36, we obtain the dynamic differential rate of ionization due to the absorption of n photons:

$$\begin{aligned} d\Gamma_\mathrm{ion}^{(n)}(t_\mathrm{d}; \boldsymbol{p}) = 2\pi |\langle \Phi_{v_0 J_0 M_0}(t_\mathrm{d}) | T_\mathrm{ion}^{(n)}(\boldsymbol{p}, \boldsymbol{R}) | \Phi_{v_0, J_0, M_0}(t_\mathrm{d}) \rangle|^2 \\ \times \delta(p^2/2 + U_p + I_p - n\omega) \frac{d^3 p}{(2\pi)^3}, \end{aligned} \tag{37}$$

where $\boldsymbol{p}$ is the momentum of the ejected electron. Summing over all allowed photon numbers $n \geq n_0 \equiv [(I_p + U_p)/\omega]_\mathrm{int.} + 1$ and taking the thermal average with respect to the Boltzmann distribution of the ro-vibrational states, we obtain the dynamic ionization signal:

$$\begin{aligned} \frac{dS_\mathrm{ion}^{(n)}(t_\mathrm{d}; \hat{\boldsymbol{p}})}{d\hat{\boldsymbol{p}}} = 2\pi \sum_{n \geq n_0} \sum_{v_0, J_0, M_0} \rho_{v_0, J_0, M_0} |\langle \Phi_{v_0 J_0 M_0}(t_\mathrm{d}) | T_\mathrm{ion}^{(n)}(\boldsymbol{p}, \boldsymbol{R})| \\ \times \Phi_{v_0, J_0, M_0}(t_\mathrm{d}) \rangle|^2 \rho(E(p_n)), \end{aligned} \tag{38}$$

where $\rho(E(p_n)) \equiv \sqrt{2(n\omega - I_p - U_p)}/(2\pi)^3$ is the density of final states per unit interval of the kinetic energy of the emitted electron due to absorption of n photons, and $d\hat{\boldsymbol{p}} = \sin\theta_p d\theta_p d\phi_p$.

4.1 A molecular clock

Below we give a simple analytic example of the excitation of coherent vibrations in a homonuclear diatomic molecule. Since the latter does not possess any permanent dipole moment, the laser–molecule interaction in the electronic ground state of the molecule is mediated by the polarizability $\alpha(R)$ alone, which might vary slowly with the internuclear separation R. Note that for a common laser frequency ω, such that the energy of the photon is smaller than the first electronic excitation energy of the molecule, $\alpha(R)$ is a positive quantity. Let the electric field of the linearly polarized incident laser pulse be given by,

$$\boldsymbol{F}(t) \equiv \boldsymbol{F}_0(t)\cos(\omega t + \delta), \tag{39}$$

where $\boldsymbol{F}_0(t)$ is the pulse envelope and δ is an initial phase (measured, e.g., from the peak of the pulse, assumed to occur at $t = 0$). The laser–molecule interaction potential can be written therefore as

$$\begin{aligned} V(R;t) &= -\frac{1}{2}\alpha(R)F^2(t) \\ &= -\frac{1}{2}\alpha(R)F_0^2(t)\cos^2(\omega t + \delta). \end{aligned} \tag{40}$$

The slow variation of $\alpha(R)$ about the equilibrium separation $R = R_e$ allows one to approximate it by a linear expansion, $\alpha(R) = \alpha(R_e) + \alpha'(R_e)(R - R_e)$ where $\alpha'(R_e)$ is its derivative at the equilibrium distance R_e. Thus, the Hamiltonian of the vibrational motion can be approximated as,

$$\begin{aligned} H(\xi,t) = &-\frac{1}{2\mu}\frac{\partial^2}{\partial\xi^2} + \frac{1}{2}\mu\omega_0^2\xi^2 - \frac{1}{2}F^2(t)\alpha(R_e) \\ &-\frac{1}{2}F^2(t)R_e\alpha'(R_e)\xi, \end{aligned} \tag{41}$$

where ξ is defined by $\xi \equiv (R - R_e)/R_e$, and $\boldsymbol{F}(t)$ is given by Eq. 39. The corresponding Schrödinger equation,

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = H(\xi,t)|\Psi(t)\rangle \tag{42}$$

can be solved in various ways (e.g., by path-integral method [26]). We note that at room temperature or below, essentially only the vibrational ground state is occupied for the lighter diatomics. We write the total wavepacket solution satisfying the initial condition, $|\Psi(t_0)\rangle \equiv |v = 0\rangle$, as:

$$|\Psi(t)\rangle = \sum_v C_v(t)|v\rangle e^{-iE_v t} e^{\frac{i}{2}\alpha(R_e)\int_{t_0}^{t} F^2(t')\mathrm{d}t'}. \tag{43}$$

We obtain the corresponding occupation amplitude

$$C_v(t) = \frac{(-i)^v}{\sqrt{v!}} e^{-\int_{t_0}^{t} \dot{u}^*(t')u(t')\mathrm{d}t'} [u(t)]^v/v!, \tag{44}$$

where

$$u(t) = \frac{i}{2}R_e\alpha'(R_e)\xi_{01}\int_{t_0}^{t} F^2(t')e^{i\omega_0 t'}\mathrm{d}t' \tag{45}$$

and ξ_{01} is the matrix element of the coordinate ξ with respect to the ground and the first excited vibrational state wavefunctions, $\phi_0(\xi) = \langle\xi|0\rangle$; $\phi_1(\xi) = \langle\xi|1\rangle$ and

$$\omega_0 = (E_1 - E_0)/\hbar \equiv \omega_{01} \tag{46}$$

is the transition frequency. We are now in a position to determine the ionization signal due to the probe pulse, including the amount of the phase shift ϕ_S to be observed in the signal. The phase shift ϕ_S of the signal is determined by the phase of the product of the complex dynamic amplitudes

$$Z(t) = C_0^*(t)C_1(t). \tag{47}$$

We substitute Eqs. 44 and 45 in Eq. 47 and note that

$$\begin{aligned} \int_{t_0}^{t} (\dot{u}^*(t')u(t') + \text{c.c.})\mathrm{d}t' &= \int_{t_0}^{t} (\dot{u}^*(t')u(t') + \dot{u}(t')u^*(t'))\mathrm{d}t' \\ &= \int_{t_0}^{t} \frac{\partial}{\partial t'}(u^*(t')u(t'))\mathrm{d}t' \\ &= (u^*(t)u(t)) \\ &= |u(t)|^2, \end{aligned} \tag{48}$$

where we have used the initial condition $u(t_0) = 0$. Thus, we have the simple expression,

$$Z(t) = e^{-|u(t)|^2}u(t). \tag{49}$$

Hence the ionization signal becomes:

$$\begin{aligned} S(t_\mathrm{d}) = &\langle v_0|T_\mathrm{ion}(R)T_\mathrm{ion}^\dagger(R)|v_0\rangle|C_0(\tau)|^2 \\ &+ \langle v_1|T_\mathrm{ion}(R)T_\mathrm{ion}^\dagger(R)|v_1\rangle|C_1(\tau)|^2 \\ &+ \{\langle v_0|T_\mathrm{ion}(R)T_\mathrm{ion}^\dagger(R)|v_1\rangle C_0^*(\tau)C_1(\tau)e^{i\omega_{01}t_\mathrm{d}} + \text{c.c.}\}, \end{aligned} \tag{50}$$

where τ is the duration of the pump pulse. This we may rewrite more suggestively as,

$$S(t_\mathrm{d}) = S_A + S_B\cos(\omega_{01}t_\mathrm{d} + \phi_S), \tag{51}$$

where S_A is the sum of the two constant terms of Eq. 50. We have expressed the third term above as $\frac{1}{2}|S_B|e^{i(\omega_{01}t_\mathrm{d}+\phi_S)}$, where the oscillation frequency $\omega_{01} = (E_1 - E_0)/\hbar$, and the amplitude $S_B = ((\mathrm{Re}(T_{01}(\tau))^2 + (\mathrm{Im}(T_{01}(\tau))^2)^{1/2}$, where $T_{01}(\tau) \equiv \langle v_0|(|T_\mathrm{ion}(R)|^2)|v_1\rangle C_0^*(\tau)C_1(\tau)$ stands for the complex pre-factor of the third term in Eq. 50. Clearly the above result predicts a coherent periodic motion of the ionization signal as a function of the time-delay t_d, with a frequency equal to the transition frequency $\omega_{01} = (E_1 - E_0)/\hbar$ between the two vibrational levels, and a finite phase-shift given by $\phi_S = \arctan(\mathrm{Im}(T_{01}(\tau))/\mathrm{Re}(T_{01}(\tau)))$. To see the close connection between the coherent ionization signal, Eq. 50 above, and the local variation of the nuclear positions driven by the ionizing field, we compute the expectation value of the deviation of the nuclear separation R whose equilibrium value is R_e. Semi-classically, we may associate with the above motion a variation of the center of the vibrational wave-packet state that could be excited by the pump pulse from the initially occupied vibrational ground state $|v = 0\rangle$.

$$\begin{aligned}\langle R\rangle(t) &= R_e + \langle\Phi_{v_0}(t)|(R-R_e)|\Phi_{v_0}(t)\rangle\\ &= R_e + R_e\langle\Phi_{v_0}(t)|(R-R_e)/R_e|\Phi_{v_0}(t)\rangle\\ &= R_e(1+|C_0(\tau)|^2\langle v_0|\xi|v_0\rangle + |C_1(\tau)|^2\langle v_1|\xi|v_1\rangle)\\ &+ R_e(\{C_0(\tau)C_1^*(\tau)\langle v_1|\xi|v_0\rangle e^{i\omega_{01}t_d} + \text{c.c.}\})\\ &= R_e(A+|B|\cos(\omega_{01}t_\mathrm{d}+\phi)) \qquad (52)\end{aligned}$$

where we have used $\langle v_1|(R-R_e)|v_0\rangle = R_e\ \langle v_1|\xi|v_0\rangle$, etc. We note that the vibrational transition matrix elements are real, and we have written the sum of the constant terms as R_eA, and the amplitude of the interference term as $\frac{1}{2}R_e|B|e^{i\phi}$, where the phase shift $\phi = \arctan(\mathrm{Im}(C_0(\tau)C_1^*(\tau))/\mathrm{Re}(C_0(\tau)C_1^*(\tau)))$. This result explicitly shows that the nuclear separation as a function of time $\langle R\rangle(t)$, t $\equiv \tau + t_\mathrm{d}$ executes a coherent periodic oscillation with a frequency equal to the vibrational excitation frequency, $\omega_{01} = (E_1 - E_0)/\hbar$. Moreover, this clock-motion shows a definite phase shift ϕ defined above. We may conclude by pointing out that when a large number of states are excited by the pump-pulse (e.g., in the case of a heavy diatomic molecule), a more involved modulation of the ionization current is to be expected that should be characterized by quasi-periodic fractional revivals of the ionization signal (analogous to the revivals in the case of the rotational coherence [9, 10]).

5 Summary

A theory of excitation of coherent ro-vibrational motions in linear molecules and their detection in dynamic HHG and ionization signals in intense-field pump–probe experiments is presented. Explicit expressions correlating the nuclear motions in real time with the modulation of the HHG and ionization signals are derived, and a simple analytic example of the excitation of the “molecular clock” motion, associated with the “0–1” vibration in a homonuclear diatomic molecule, and its detection in the ionization signal, is given.

References

1. Kanai T, Minemoto S, Sakai H (2005) Nature 435:470
2. Baker S, Robinson JS, Haworth CA, Teng H, Smith RA, Chirila CC, Lein M, Tisch JWG, Marangos JP (2006) Science 312:424
3. Corkum PB, Krausz F (2007) Nature Phys 3:381
4. Torres R, Kajumba N, Underwood JG, Robinson JS, Baker S, Tisch JWG, de Nalda R, Bryan WA, Velotta R, Altucci C, Turco ICE, Marangos JP (2007) Phys Rev Lett 98:203007
5. Kajumba N, Torres R, Baker S, Robinson JS, Tisch JWG, Marangos JP, Underwood JG, de Nalda R, Altucci C, Velotta R, Bryan WA, Turcu ICE et al (2006) Central laser facility annual report 2005/2006V. Rutherford Appleton Laboratory, UK, p 80
6. Yoshii K, Miyaji G, Miyazaki K (2008) Phys Rev Lett 101:183902
7. Lein M (2007) J Phys B 40:R135
8. Faisal FHM, Abdurrouf A, Miyazaki K, Miyaji G (2007) Phys Rev Lett 98:143001
9. Faisal FHM, Abdurrouf A (2008) Phys Rev Lett 100:123005
10. Abdurrouf A, Faisal FHM (2009) Phys Rev A 79:023405
11. Bandrauk AD, Chelkowski S, Kawai S, Lu H (2008) Phys Rev Lett 101:153901
12. Bandrauk AD, Chelkowski S, Lu H (2009) J Phys B 42:075602
13. Becker A, Faisal FHM (2005) J Phys B 38:R1–R56
14. Muth-Böhm J, Becker A, Faisal FHM (2000) Phys Rev Lett 85:2280, Erratum: Phys Rev Lett 96:039902 (2006)
15. Becker A, Faisal FHM (1996) J Phys B 29:L197
16. Becker A, Faisal FHM (2000) Phys Rev Lett 84:3546
17. Lewenstein M, Balcou Ph, Ivanov MYu, L’Huillier A, Corkum PB (1994) Phys Rev A 49:2117
18. Corkum PB (1993) Phys Rev Lett 71:1994
19. Kulander KC, Schafer KJ, Krause JL (1994) In: Piraux B, L’Huillier A, Rzazewski K (eds) Super-intense laser-atom physics, NATO ASI Ser B, vol 316. Plenum Press, New York, p 95
20. Feynman RP (1962) Quantum electrodynamics. Benjamin Inc, New York
21. Faisal FHM (1987) Theory of multiphoton processes. Plenum Press, New York
22. Faisal FHM, Schlegel G (2005) J Phys B 38:L223
23. Chase DM (1956) Phys Rev 104:838
24. Faisal FHM, Temkin A (1972) Phys Rev Lett 28:203, Erratum: Phys Rev Lett 29:320 (1972)
25. Schmidt MW, Baldridge KK, Boatz JA, Elbert ST, Gordon MS, Jensen JH, Koseki S, Matsunaga N, Nguyen KA, Su S, Windus TL, Dupuis M, Montgomery JA Jr (1993) J Comput Chem 14:1347
26. Feynman RP, Hibbs AR (1965) Quantum mechanics and path integrals. McGraw-Hill, New York