

High-order-harmonic generation in dimensionally reduced systemsGiuseppe Castiglia, Pietro Paolo Corso, Dario Cricchio, Rosalba Daniele, Emilio Fiordilino,^{*}
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The time-dependent wave function of a nanoring driven by a laser field is obtained by exploiting the symmetries inherent to the system and used for studying the properties of the electromagnetic radiation emitted by the nanoring as a function of the polarization state of the laser. The diffused radiation has the characteristics of high-order-harmonic generation. For a noncircularly polarized laser field an extension of the expected cutoff position is evident, indicating that nanorings are efficient sources of radiation. The polarization state of the emitted harmonics can be opportunely controlled by varying the parameters of the pump field. The profile of the absorbed angular momentum shows that a magnetic moment can be induced depending on the polarization of the driven field.

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I. INTRODUCTION

Mesoscopic materials extending along one or two spatial dimensions for several orders of magnitude more than the other(s) are now fabricated and accessible to experimental research. Among these new materials we mention thin rings [1], dots, and molecules of dots [2]. From the quantum point of view, they provide the researcher the unique possibility of penetrating the nature of objects that can be described as reduced dimensionality systems (RDS). The treatment of such a system is made possible by the fact that the gap between the energy levels pertaining to the dynamics of the electrons along the small dimension is very large; since these degrees of freedom are frozen and unavailable to the electrons, they can be essentially neglected in the theoretical problem.

Recently it has been shown that carbon can be found in allotrope states describable as RDS. Graphene is a monolayer of carbon atoms, situated on the vertices of hexagons, continuously covering a planar surface like a honeycomb. The substitution of particular hexagons with pentagons bends the planar structure to form hollow cages, called fullerenes or also buckyballs; among these the C₆₀ molecule is mostly famous: An impressive distribution of regular hexagons and pentagons gifts it of a suggestive spherical shape, with radius 6.7 a_0 , resembling a soccer ball (a_0 is the Bohr radius and the natural length unit in atomic and molecular studies). The graphene sheet can also be rolled to form single- or double-walled nanotubes and cones (nanohorns, the last synthesized carbon allotrope).

All of these carbon molecules are truly members of the RDS [3]. Let us consider, for example, a nanotube of length L and radius R . The energy levels of one electron of mass m_e constrained to move on the surface are given by the expression,

$$E_{n,m} = \frac{\hbar^2}{2m_e} \left(\frac{\pi^2 n^2}{L^2} + \frac{m^2}{R^2} \right), \quad (1)$$

with $n > 0$ and $m \geq 0$ integers. The fabrication of single-walled nanotubes has been reported [4] with $\pi^2 R^2/L^2 \approx 10^{-15}$; the population of energy levels with $m > 0$ is possible only when states with $n \gg 1$ are filled. It is therefore perfectly

legitimate to approximate the nanotubes as one-dimensional wires.

Historically, an interesting parallelism can be found in the Kaluza-Klein cosmology that discusses a model of universe with four spatial dimensions; the extra coordinate, however, is compact, ranges in a very small interval of values, and, therefore, is essentially unpopulated by quantum particles. Although it does not seem likely that studies of mesoscopic matter will ever need more than three spatial coordinates per particle, it is suggestive that quantum physics makes experimentally possible the investigation of systems with different dimensions and the incoming of new effects when physical parameters makes possible the transition to a different dimensionality.

In atoms or molecules interacting with an intense electromagnetic field, the consideration that the dynamics of the active electrons is essentially determined by the laser electric field makes it advantageous to use a single spatial dimension for calculating the desired quantities such as high-harmonic spectra or single- or double-ionization probability [5–9].

Moreover, the use of a reduced dimensionality model often does not hide the effects and permits clear vision of the physics; in this class of problems the Kronig-Penney model, which unveiled the band structure of conductors, deserves particular mention [10].

From the mathematical point of view, the Schrödinger equation of an electron bound to a curved surface contains additional, geometry-induced, gauge potentials which make the discussion of the problem less obvious and more interesting than expected; a very short account of the procedures is given in [11] but the interested reader will find a thorough treatment in [12,13]; the theory therefore can be applied to geometries such as the Moebius surface [14].

The large number of atoms and their position within the mentioned carbon allotropes can result in a simplification of the theoretical treatment. Actually relevant information on the physics of the systems can be gained by judicious use of the inherent symmetries of the problem. In Ref. [11] the quasispherical symmetry of buckyballs has been exploited by using a multielectron two-dimensional jellium model to obtain a wealth of information on the structure and stability of these

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elements which has been compared with experimental data. In Ref. [15] a three-dimensional jellium model and density functional theory were used to calculate the photoionization cross section of C₆₀. In Ref. [16] the treatment has been generalized to obtain the analytical wave function of one electron on a spherical segment enclosed by radial impenetrable walls.

In dealing with nanostructures driven by strong electromagnetic fields, the exploitation of the symmetries results in a relevant simplification of the problem at hand which seems not to hide the essence of the problem and unveils the origin of laser-induced effects that could be easily hidden in more complex treatments.

For example, in [17,18] it is shown that unipolar electromagnetic pulses driving a thin ring can induce an oscillating dipole moment with subsequent emission of electromagnetic field; similar equations are found in the study of alignment of rotators by laser pulses [19]. The presence of a static magnetic field does indeed modify the charge polarization and the ensued emitted light [20]. In [21] the response of a ring to laser pulses is investigated. An interesting debate on the role played by dynamical symmetries of order N and how to exploit them can be found in [22–26].

The interaction of a strong laser pulse of angular frequency ω_L with atoms and molecules gives the opportunity to observe processes which are nonlinear in the laser intensity and cannot be described by the standard perturbation theory. Among these processes we mention multiphoton ionization [27], above-threshold ionization [28], high-harmonic generation [29], above-threshold dissociation [30,31], and nonsequential multiple ionization of atoms and molecules [7,32–34].

Within the scope of this paper we are particularly interested in high-order-harmonic generation (HHG) which is one of the most studied effects in the physics of laser-matter interaction; it consists of the diffusion by laser driven atoms, molecules, and nanoparticles of high harmonics of the pumping field. The origin of the radiation resides in the highly nonlinear modification of the electron wave function induced by the pump that, in turn, results in a broad Fourier spectrum consisting of odd multiples of the fundamental laser frequency from atoms and of odd and, occasionally, even harmonics from molecules. Lately few analytical and experimental studies have shown that the large sizes and polarizability makes mesoscopic molecules interesting sources of harmonics [11,22,23,25,26,35–37]. Among carbon allotropes, the C₆₀ has deserved attention and ingenuity in devising experimental and theoretical models [38–45]; it has been shown that the use of C₆₀ can also produce short pulses with duration of the order of $\tau \approx 2\sqrt{m_e R/e\mathcal{E}_L}$ with R the radius of the fullerene, \mathcal{E}_L the laser electric field strength, and $-e$ the electron charge [42].

This paper deals with the response of a nanoring driven by a laser field in different states of polarization with particular attention to the spectrum emitted by the laser-induced charge oscillations. Adopting all the possible simplifications that do not hide the essential effects, we consider a single active electron system, bound on a circumference, driven by a laser electric field lying in the plane of the ring. Our model, therefore, is in the framework of RDS. We calculate the spectrum emitted by the electron, the energy and the angular moment transferred by the laser to the electron, and the parameters characterizing the polarization state of the

harmonics. The diameter of the nanoring is equal to the radius of the hexagonal cell of the graphene. We show that a current can be induced to flow along the ring resulting in a quantum averaged current and magnetic moment. It is not hazardous to suppose that a graphene sheet, built up of many elemental cells, under the influence of a weak laser field might be circulated by an electric current. The large geometrical size of the structure could become a relevant emitter of harmonics.

II. THEORY

Let us consider an electron constrained over a circle of radius R lying in the x - y plane and acted upon by a laser elliptically polarized in the same plane.

The number of free parameters entering the problem is large and might entwine the relation between causes and effects into a Gordian knot; thus, in the following, we undertake all the simplifying steps which can help the comprehension. With this guiding idea we confine our treatment to relatively long pulse duration in order to reduce effects related to envelope phase and line broadening. The external electric field is therefore taken of the form,

$$\mathcal{E}(\mathbf{r}, t) = \mathcal{E}_L f(t) [\boldsymbol{\epsilon}_x \cos \beta \cos(\omega_L t) + \boldsymbol{\epsilon}_y \sin \beta \sin(\omega_L t)], \quad (2)$$

$\boldsymbol{\epsilon}_x$ and $\boldsymbol{\epsilon}_y$ being the unit vectors along the x and y axes respectively, and $\beta \in [0, \pi/2]$ a parameter characterizing the field polarization: $\beta = 0$ gives a laser polarized along the x axis, $\beta = \pi/2$ gives a laser polarized along the y axis, $\beta = \pi/4$ gives a circular polarization. It is worth noting that the phase of the field, for linear polarization along the two axes, differs by $\pi/2$, therefore slightly different results are to be expected in the two cases. The pulse shape of the field is described by the function $f(t)$.

We solve the time-dependent Schrödinger equation,

$$i\hbar \frac{\partial}{\partial t} |t\rangle = \hat{H} |t\rangle, \quad (3)$$

with

$$\hat{H} = \hat{H}_0 + \hbar\Omega(t) [\cos \beta \cos(\omega_L t) \cos \varphi + \sin \beta \sin(\omega_L t) \sin \varphi], \quad (4)$$

the full time-dependent Hamiltonian of the problem. Here,

$$\hat{H}_0 = \frac{\hbar^2}{2m_e R^2} \hat{\ell}_z^2 \quad (5)$$

is the laser free Hamiltonian, with only the kinetic energy term, and

$$\hbar\Omega(t) \equiv e\mathcal{E}_L R f(t) \quad (6)$$

describes the laser-ring maximum interaction energy; $\hat{\ell}_z$ is the usual z component of the orbital angular momentum operator (in units of \hbar) whose eigenvectors and eigenvalues are the well-known angular momentum states,

$$\hat{\ell}_z |m\rangle = m|m\rangle, \quad m = 0, \pm 1, \pm 2, \dots; \quad (7)$$

the analytical expression for $|m\rangle$ being

$$|m\rangle \rightarrow \Phi_m(\varphi) = \frac{e^{im\varphi}}{\sqrt{2\pi}}. \quad (8)$$

The states $|m\rangle$ are eigenstates of \hat{H}_0 with energy,

$$\hat{H}_0|m\rangle = \hbar\omega_m|m\rangle \Rightarrow \hbar\omega_m = \frac{\hbar^2 m^2}{2m_e R^2}, \quad (9)$$

and provide a suitable basis for the Hilbert space of our problem. The ground state $|0\rangle$ is not degenerate while the states $|\pm m\rangle$ have the same energy.

We expand the time-dependent state $|t\rangle$ as a linear combination of the bare states,

$$|t\rangle = \sum_{m=-\infty}^{+\infty} a_m(t)|m\rangle, \quad (10)$$

by making use of the matrix elements,

$$\langle n|\cos\varphi|m\rangle = \frac{1}{2}(\delta_{m-n+1,0} + \delta_{m-n-1,0}), \quad (11)$$

$$\langle n|\sin\varphi|m\rangle = \frac{-i}{2}(\delta_{m-n+1,0} - \delta_{m-n-1,0}), \quad (12)$$

we obtain a set of coupled equations for the probability amplitudes:

$$i\dot{a}_n = \omega_n a_n + \frac{\Omega(t)}{2} \{ [\cos\beta \cos(\omega_L t) - i \sin\beta \sin(\omega_L t)] a_{n-1} + [\cos\beta \cos(\omega_L t) + i \sin\beta \sin(\omega_L t)] a_{n+1} \}, \quad (13)$$

showing a ladder type coupling of the states; this set of equations has been solved by numerical integration. The gap between the energy levels increases linearly with $|n|$ and we expect that upper states are negligibly populated and do not contribute to the evolution of the systems. Therefore we solve this set of coupled equations by mere truncating the system for $|m| \geq 10$; we checked that states with $|m| > 10$ do not contribute to the final result for the values of the physical parameters we use in this paper.

From the state $|t\rangle$ we can calculate all time-dependent quantum averaged (TDQA) parameters that are relevant to our ends. The TDQA dipole moment is $\mathbf{D}(t) = -e\mathbf{r}(t)$, with

$$\begin{aligned} \mathbf{r} &= \epsilon_x \langle t|x|t\rangle + \epsilon_y \langle t|y|t\rangle \\ &= \sum_{n=-\infty}^{+\infty} [\epsilon_x \text{Re}(a_{n-1}^* a_n) + \epsilon_y \text{Im}(a_n^* a_{n-1})] \end{aligned} \quad (14)$$

the TDQA position of the electron, the TDQA energy being given by

$$E(t) = \sum_{m=-\infty}^{+\infty} |a_m(t)|^2 \hbar\omega_m, \quad (15)$$

and the TDQA angular momentum acquired by the electron being

$$L_z(t) = \sum_{m=-\infty}^{+\infty} |a_m(t)|^2 m\hbar. \quad (16)$$

To these quantities we associate the correspondent time-independent quantum averaged quantities:

$$\langle E \rangle = \frac{1}{T} \int_0^T E(t) dt, \quad (17)$$

and

$$\langle L_z \rangle = \frac{1}{T} \int_0^T L_z(t) dt, \quad (18)$$

with T the laser pulse duration.

In the last two decades, HHG attracted much attention because it opens the route towards devices generating high frequency and coherent electromagnetic radiation. In atoms driven by a linearly polarized field, it already seems a well-established fact that the maximum obtainable frequency ω_M (cutoff) is given by the general law,

$$\hbar\omega_M \cong |W| + 3U_p = |W| + 3 \frac{e^2 \mathcal{E}_L^2}{4m_e \omega_L^2}, \quad (19)$$

with W the ground-state energy and U_p the ponderomotive potential.

The relation (19) can be easily determined from mere energy conservation law and classical physics; it corresponds to a classical trajectory of the electron that reaches the continuum and then is recaptured by the parent ion [33,46]; such a trajectory exists only for a linearly polarized laser field. All the mechanism is dubbed three-step model; during the first step the laser field extracts the active electron out of a bound state to the continuum with zero kinetic energy. Then the electron drifts along the direction of the laser electric field gaining energy (second step). When the laser electric field reverses its direction, the electron is accelerated back towards the atomic or molecular parent ion; during this recollision the electron, if it recombines into the bound state, releases the energy acquired during the round trip emitting the cutoff energy in an abrupt collision (third step). By quantal or classical calculations the favorable trajectory that gives rise to the emission of $\hbar\omega_M$ can be determined [47]. At this point it should be stressed that the cutoff photon is emitted during the head-on collision and recapture process but any electromagnetic emission is essentially due to the acceleration of the electron. HHG has been obtained for physical situations when no recollision is presented such as two-level systems [48–50] or even when no bound state is supported such as repulsive potentials [51]. Other cutoff laws have been obtained for two-level atoms or molecules [50,52–54].

If a source of high frequency radiation is looked after, it is of paramount importance to be able to increase the value of ω_M by changing the free physical parameters. However, the rigid law in Eq. (19) seems to leave to the researcher the laser parameters and, essentially, \mathcal{E}_L/ω_L as efficient control knob.

Sources other than atoms are under investigation. Among the more promising there is to consider molecules providing all their free parameters as control knobs. According to theory, the emission power proportional to $|\mathbf{r}(\omega)|^2$ ($\mathbf{r}(\omega)$ is the Fourier transform of $\mathbf{r}(t)$); molecules have larger sizes, are more polarizable than atoms, and, thus, can give a good harmonic yield at relative lower laser intensity. Nanomolecules have large dimensions, display simplifying symmetries that paradoxically can make calculations easier than for smaller molecules, and, when nanodots are at hand, can be tailored at will.

One of the points of interest of this paper is the study of the dependence of the polarization state of the emitted harmonics upon the polarization state of the pump laser. Elliptically

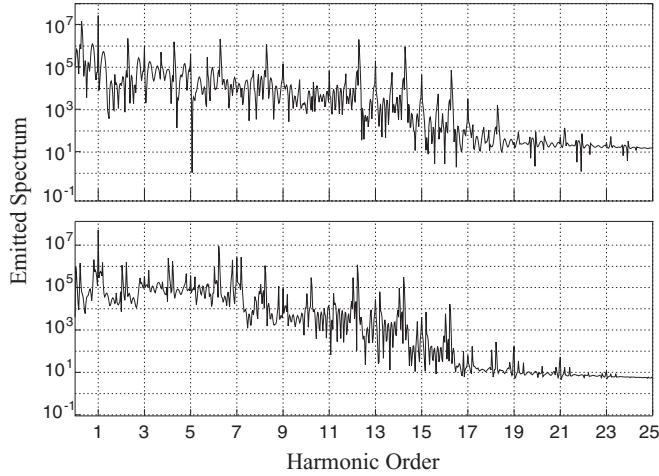


FIG. 1. Spectrum emitted by the nanoring. The relevant parameters entering the calculations are $I_L = 4 \times 10^{14} \text{ W cm}^{-2}$, $\lambda_L = 1969 \text{ nm}$ ($\hbar\omega_L = 0.63 \text{ eV}$). (Top) $\beta = 0^\circ$ (laser polarized along the x axis); (bottom) $\beta = 20^\circ$ (elliptically polarized laser, $I_x = 3.5 \times 10^{14} \text{ W cm}^{-2}$; $I_y = 4.7 \times 10^{13} \text{ W cm}^{-2}$).

polarizable laser fields, that do not favor HHG from atoms, can stimulate emission from molecules; this is an appealing feature for molecules since it allows the control of the polarization of the harmonics [55]. Moreover nanorings can be magnetized and the question is still a scarcely explored ocean.

All of this suggests the study of a nanoring driven by a laser field in different polarization states. Again, to keep the extension of the paper within reasonable boundaries, we shall concentrate solely on the main effects without following all the rivulets furrowing the area whose exploration is deferred to subsequent papers.

III. RESULTS

Here we present calculations of a nanoring with radius $R = 2.7a_0$ driven by a laser field given in Eq. (2); $f(t)$ is a

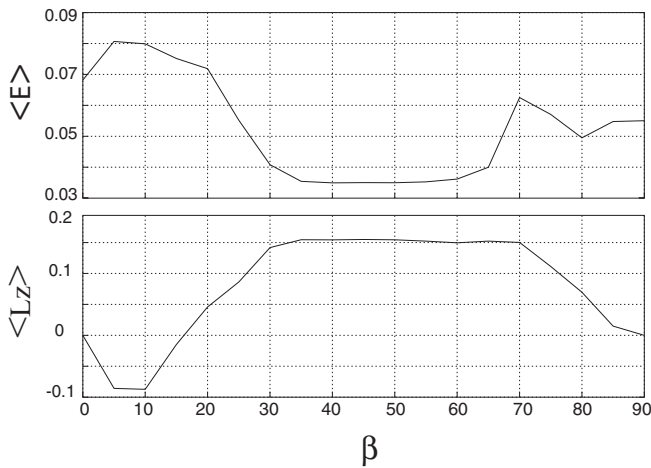


FIG. 2. (Top) Average energy (in au); (bottom) average angular momentum (in units of \hbar) absorbed by the ring in the whole laser shot versus the polarization parameter β in degrees. The relevant parameters used for the calculations are $I_L = 4 \times 10^{14} \text{ W cm}^{-2}$, $\lambda_L = 1969 \text{ nm}$ ($\hbar\omega_L = 0.63 \text{ eV}$).

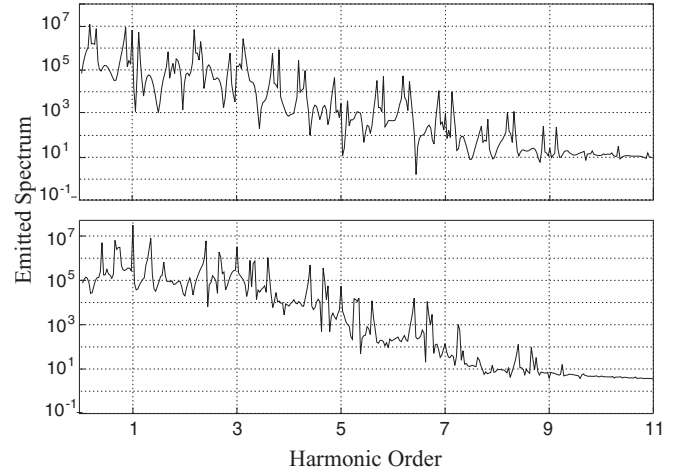


FIG. 3. Spectrum emitted by the nanoring. The relevant parameters used for the calculations are $I_L = 4 \times 10^{14} \text{ W cm}^{-2}$, $\lambda_L = 591 \text{ nm}$ ($\hbar\omega_L = 2.1 \text{ eV}$). (Top) $\beta = 0^\circ$ (laser polarized along the x axis); (bottom) $\beta = 20^\circ$ (elliptically polarized laser, $I_x = 3.5 \times 10^{14} \text{ W cm}^{-2}$; $I_y = 4.7 \times 10^{13} \text{ W cm}^{-2}$).

trapezoidal function with ramps lasting 4 optical cycles (oc) and total duration of 32 oc. Since we are interested in the response of the system to different laser polarization, we decide to keep the overall laser intensity $I_L \propto \mathcal{E}_L^2 = \mathcal{E}_x^2 + \mathcal{E}_y^2$ constant. Here, according to Eq. (2), $\mathcal{E}_x = \mathcal{E}_L \cos \beta$ and $\mathcal{E}_y = \mathcal{E}_L \sin \beta$ denote the maximum values of the driving field along the x and y directions, respectively. As a rule we denote the laser with I_L given in W cm^{-2} .

The polarization of the n th harmonic field is determined by the angle ψ and the eccentricity e_c defined as [55,56]

$$\tan 2\psi = \tan(2\alpha) \cos \delta, \quad 0 \leq \psi < \pi, \quad (20)$$

$$e_c = \tan \chi, \quad -\pi/4 < \chi \leq \pi/4, \quad (21)$$

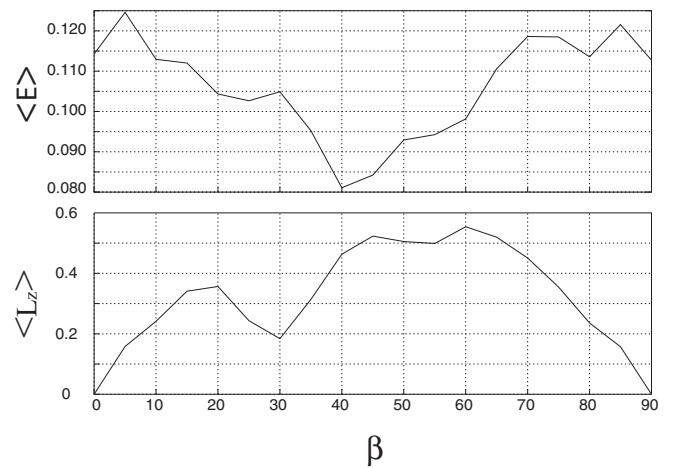


FIG. 4. (Top) Average energy (in au); (bottom) average angular momentum (in units of \hbar) absorbed by the ring in the whole laser shot versus the polarization parameter β in degrees. The relevant parameters used for the calculations are $I_L = 4 \times 10^{14} \text{ W cm}^{-2}$, $\lambda_L = 591 \text{ nm}$ ($\hbar\omega_L = 2.1 \text{ eV}$).

TABLE I. Polarization angle ψ [up value in $\binom{0}{0}$] and eccentricity e_c [bottom value in $\binom{0}{0}$] versus the order n of the harmonic for different value of the parameter β . The relevant parameters of the calculations are $\lambda_L = 591$ nm ($\hbar\omega_L = 2.1$ eV), $I_L = 4 \times 10^{14}$ W cm $^{-2}$. The not given values are for harmonics not present in the spectrum.

$\beta \downarrow n \rightarrow$	1	3	5	7	9	11	13	15	17	19	21
0	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{-}{0}$	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{0}{0}$	$\binom{-}{0}$	$\binom{-}{0}$
5	$\binom{0}{0.54}$	$\binom{-}{0}$	$\binom{90}{0.12}$	$\binom{89}{-0.1}$	$\binom{103}{-0.36}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{3}{0.45}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$
10	$\binom{1}{0.69}$	$\binom{5}{-0.16}$	$\binom{94}{0.09}$	$\binom{91}{0.28}$	$\binom{88}{0.13}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{0}{-0.36}$	$\binom{-}{0}$	$\binom{-}{0}$
15	$\binom{2}{0.6}$	$\binom{-}{0}$	$\binom{101}{0.64}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{20}{0.25}$	$\binom{4}{0.6}$	$\binom{124}{0.62}$	$\binom{72}{0.61}$	$\binom{50}{0.54}$
20	$\binom{1}{0.65}$	$\binom{13}{-0.28}$	$\binom{176}{-0.76}$	$\binom{86}{0.5}$	$\binom{78}{-0.45}$	$\binom{-}{0}$	$\binom{178}{-0.19}$	$\binom{2}{0.25}$	$\binom{159}{-0.48}$	$\binom{99}{-0.08}$	$\binom{99}{-0.24}$
25	$\binom{1}{0.83}$	$\binom{1}{0.76}$	$\binom{165}{-0.01}$	$\binom{89}{0.5}$	$\binom{106}{0.08}$	$\binom{-}{0}$	$\binom{3}{0.36}$	$\binom{5}{0.56}$	$\binom{20}{0.7}$	$\binom{65}{-0.05}$	$\binom{60}{-0.43}$
30	$\binom{1}{0.93}$	$\binom{8}{0.9}$	$\binom{175}{0.47}$	$\binom{87}{0.63}$	$\binom{99}{-0.42}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{17}{0.12}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$
35	$\binom{0}{0.97}$	$\binom{1}{0.98}$	$\binom{172}{0.45}$	$\binom{88}{0.88}$	$\binom{96}{0.95}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$
40	$\binom{0}{0.99}$	$\binom{4}{1}$	$\binom{5}{0.82}$	$\binom{43}{0.85}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$
45	$\binom{103}{1}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$	$\binom{-}{0}$

with $\sin 2\chi = \sin 2\alpha \sin \delta$, $\tan \alpha = (\mathcal{E}_x^{(n)}/\mathcal{E}_y^{(n)})$, and δ the relative phase between $\mathcal{E}_x^{(n)}$ and $\mathcal{E}_y^{(n)}$; the negative (positive) sign for e_c takes into account the clockwise (counterclockwise) orientation of the emitted harmonic.

We discuss the response of the system to a laser operating in the infrared and visible laser regime and start our discussion with the case when $\hbar\omega_L = 0.63$ eV so that $\omega_1 - \omega_0 = 3\omega_L$. The laser intensity is taken as $I_L = 4 \times 10^{14}$ W cm $^{-2}$. With such a choice of the parameters, Eq. (19) gives an expected cutoff of the emission in atoms at $\omega_M = 12\omega_L$.

In Fig. 1 we show the spectrum emitted by the nanoring for two different polarizations. A circularly polarized pump does not produce harmonics and this is to be waited for, since the model is too symmetric to give a significant yield in the actual situation. Movies of the behavior of the electronic packet show the presence of a rotating packet. Linear and elliptic polarization show emission lines (harmonic and not) and an extended cutoff.

Always for the same laser intensity I_L in Fig. 2 we show the absorbed energy $\langle E \rangle$ [from Eq. (17)] and the acquired angular momentum $\langle L_z \rangle$ [from Eq. (18)]: There is a net absorption of energy and angular momentum during the laser shot at all values of β . Letting aside the obvious result for linear polarization, we see that for a wide range of angles the transferred momentum is distributed along a plateau. This means that a magnetic momentum can be induced in a nanoring by a loosely polarized laser field.

We have performed similar calculations for $\hbar\omega_L = 2.1$ eV and the same value of the laser intensity giving a nominal cutoff energy $\omega_M = 3\omega_L$. In Fig. 3 the spectra are shown. Again the cutoff is more extended than expected and shows that nanorings are efficient emitters. This appealing result can be motivated by considering the differences between atoms and nanorings. In atoms, by using the three-step model, the cutoff is the maximum energy obtainable by the active electron and

compatible with a recapture act. Conceptually in nanorings there is no upper limit to the energy that can be gained by the electron and delivered in a single photon emission. It is surprising that a simple system with states coupled in a ladder way can still emit all the energy in a single shot. Of course the position of the plateau is an important parameter both for fundamental and technological reasons. Its analytical determination has been shown to be laborious in the simple two-level system [50]; in nanorings its position and extension is intriguing but as yet unexplained.

Figure 4 shows the averaged energy $\langle E \rangle$ absorbed by the ring and the averaged absorbed angular momentum $\langle L_z \rangle$. Again the previously shown trend is confirmed although the averaged absorbed angular momentum does not show the previous plateau as a function of the polarization parameter β .

One of the most interesting uses of HHG is the possibility of creating very short, isolated, pulses. To this goal several schemes have been developed; particularly attractive seems the use a driving laser field of time-dependent polarization [57,58]. In Tables I and II we list the polarization angles ψ and the eccentricity e_c versus the order of the emitted harmonic. It is clear from the data that the polarization state of any harmonic is controlled by the polarization state of the pumping field. This feature can be of great use in the synthesis of short harmonics via the control of the polarization state of the pump field. Preliminary results show that the harmonic radiation of the nanoring can be synthesized to give pulses of duration $\sim 1/10$ oc.

The described effects are robust against modification of duration and shape of the pulse. We obtained the response of the nanoring to trapezoidal pulses with duration 16 and 64 oc and checked that the spectrum is essentially unchanged by this parameter. Instead the use of a Gaussian or \sin^2 pulse with 32 oc makes the spectrum regularly noisier.

TABLE II. Polarization angle ψ [top value in $\begin{pmatrix} 0 \\ 0 \end{pmatrix}$] and eccentricity e_c [bottom value in $\begin{pmatrix} 0 \\ 0 \end{pmatrix}$] versus the order n of the harmonic for different value of the parameter β . The relevant parameters of the calculations are $\lambda_L = 591$ nm ($\hbar\omega_L = 2.1$ eV), $I_L = 4 \times 10^{14}$ W cm $^{-2}$. The not given values are for harmonics not present in the spectrum.

$\beta \downarrow n \rightarrow$	1	3	5	7	9
0	$\begin{pmatrix} 0 \\ 0 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} 0 \\ 0 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
5	$\begin{pmatrix} 6 \\ 0.92 \end{pmatrix}$	$\begin{pmatrix} 87 \\ 0 \end{pmatrix}$	$\begin{pmatrix} 111 \\ 0.78 \end{pmatrix}$	$\begin{pmatrix} 163 \\ 0.6 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
10	$\begin{pmatrix} 76 \\ 0.99 \end{pmatrix}$	$\begin{pmatrix} 81 \\ -0.35 \end{pmatrix}$	$\begin{pmatrix} 88 \\ 0.73 \end{pmatrix}$	$\begin{pmatrix} 26 \\ 0.99 \end{pmatrix}$	$\begin{pmatrix} 37 \\ 0.77 \end{pmatrix}$
15	$\begin{pmatrix} 87 \\ 0.9 \end{pmatrix}$	$\begin{pmatrix} 87 \\ 0.52 \end{pmatrix}$	$\begin{pmatrix} 90 \\ 0.91 \end{pmatrix}$	$\begin{pmatrix} 144 \\ 1 \end{pmatrix}$	$\begin{pmatrix} 51 \\ 0.96 \end{pmatrix}$
20	$\begin{pmatrix} 89 \\ 0.77 \end{pmatrix}$	$\begin{pmatrix} 86 \\ 0.97 \end{pmatrix}$	$\begin{pmatrix} 6 \\ 0.95 \end{pmatrix}$	$\begin{pmatrix} 160 \\ 0.93 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
25	$\begin{pmatrix} 91 \\ 0.43 \end{pmatrix}$	$\begin{pmatrix} 76 \\ 1 \end{pmatrix}$	$\begin{pmatrix} 86 \\ 0.16 \end{pmatrix}$	$\begin{pmatrix} 16 \\ 0.93 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
30	$\begin{pmatrix} 1 \\ -0.62 \end{pmatrix}$	$\begin{pmatrix} 4 \\ 0.88 \end{pmatrix}$	$\begin{pmatrix} 93 \\ 0.88 \end{pmatrix}$	$\begin{pmatrix} 150 \\ 0.96 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
35	$\begin{pmatrix} 91 \\ 0.08 \end{pmatrix}$	$\begin{pmatrix} 92 \\ 0.89 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
40	$\begin{pmatrix} 89 \\ 0.96 \end{pmatrix}$	$\begin{pmatrix} 92 \\ 1 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$
45	$\begin{pmatrix} 153 \\ 1 \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$	$\begin{pmatrix} - \\ - \end{pmatrix}$

IV. CONCLUSION

Nanoparticles such as buckyballs, nanotubes, and rings are molecules of, possibly, large dimensions. Their shape presents interesting symmetries that can be exploited to obtain information on their behavior; these symmetries are particularly at hand when dealing with a laser illuminating the material; actually it seems that the field can be as strong as to make not useful a detailed knowledge of the bare states of the molecules. The impressive simplicity of treatment possible for buckyballs and rings [11, 17] shows the opportunity to seek models giving

a quick crop of reliable information; the comparison with experiments [45] comforts and strengthens the hopes.

Rings driven by a laser can efficiently emit an electromagnetic field endowed with interesting properties. We have seen, first of all, that the spectrum of the emission is wider than expected from atoms and that the cutoff law is quite similar to the cutoff law for buckyballs; then, we have seen that the characteristics of the harmonic field can be controlled by changing the laser parameters. In particular, our investigations show that the polarization of the driving laser provides a fine tool of control; as a result, the polarization of the diffused harmonics can be tuned.

Preliminary results show that our model nanorings are suitable to emit a train of short electromagnetic pulses obeying a scaling law for the duration similar to the one for buckyballs; our simulations reveal a peculiar dependence of the train upon the laser polarization that cannot be reported here to keep the paper within a reasonable limit. An interesting point is the fact that nonzero angular momentum can be stored in the ring; a point to be developed in future research is the determination of the optimal pulse profile to maximize the storage. Since the equation of motion of the electron in the nanoring and of a rigid rotator are the same, the output from these studies can cast light on the problem of setting in motion a molecule with a definite angular velocity.

Experiments have been carried out on the second harmonic generation of a thin silver sheet [59] with a matrix of geometrical nanoholes. A small area of graphene can be seen as a collection of holes or of tangent nanorings and therefore our model calculations receive from these results confirmation and impetus.

We have elected to describe the laser pulse as a superposition of two orthogonal pulses of the same shape $f(t)$ [Eq. (2)] but, of course, the time-dependent polarization state of the driver opens a wide range of possibilities, all foreshadowing interesting channels to be explored.

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