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Signatures of the molecular potential in the ellipticity of high-order harmonics from aligned molecules

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Abstract

We explore the information content of the polarization of high-order harmonics emitted from aligned molecules driven by a linearly polarized field. The study builds upon our previous work (Ramakrishna *et al.*, Phys. Rev. A **81**, 021802(R) (2010)), which illustrated that the phase of the continuum electronic wavefunction, and hence the underlying molecular potential, is responsible, at least in part, for the ellipticity observed in harmonic spectra. We use a simple model potential and systematically vary the potential parameters to investigate the sense in which, and the degree to which, the shape of the molecular potential is imprinted onto the polarization of the emitted harmonics. Strong ellipticity is observed over a wide range of potential parameters, suggesting that the emission of elliptically polarized harmonics is a general phenomenon, yet qualitatively determined by the molecular properties. The sensitivity of the ellipticity to the model parameters invites the use of ellipticity measurements as a probe of the continuum wavefunction and the underlying molecular potential.

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I. INTRODUCTION

High-order harmonic generation (HHG)[1] is a highly nonlinear process wherein an atomic or molecular medium subject to an intense, low-frequency laser pulse emits coherent radiation with frequencies of odd-integer multiples of the driving field. The most familiar mechanism of HHG can be understood, at least at a qualitative level, in terms of a 3-step model,[2, 3] wherein the distortion of the bound state potential by the strong-field allows tunnel ionization of an electron, which is next accelerated by the laser field, first away from and next toward the core. Finally, upon recollision with the parent ion core, the driven electron may recombine, with a consequent emission of odd-harmonics of the incident frequency.

Fueled by the promise of attosecond XUV pulses, interest in HHG has been relentless since the first experiments in rare gases,[4] the early models of Refs. 2 and 5, and the numerical work of Refs. 6, 7. After the realization and characterization of trains of attosecond pulses from gaseous argon [8], the advent of experiments[9] from non-adiabatically aligned molecules [10] opened a new avenue of exciting fundamental research. In addition to possibilities of using the aligned molecular medium as a means of controlling the emitted pulses, the interest in the information content of the signal regarding the underlying electronic structure and dynamics has been inspiring.

Non-adiabatic laser alignment[10] is a coherent, strong-field approach, wherein a short (with respect to the rotational periods), moderately intense (below off-resonance ionization thresholds) laser pulse populates a rotationally broad, wave packet via sequential, Rabi-type cycles in which angular momentum is not conserved.[11] Most experiments are conducted at far-off-resonance frequencies, in which case the rotational wave packet is excited via sequential Raman cycles, with an exchange of $|\Delta J| = 0, 2$ between the field and the molecule in each cycle. The phase relations established by the sequential excitation process guarantee that the wave packet will align upon turn-off of the laser pulse, delayed with respect to the pulse peak to a degree that depends on the pulse duration and intensity, and will subsequently exhibit a coherent revival structure. In the case of linear and symmetric top molecules, the alignment established upon turn-off is precisely reconstructed periodically in time as long as coherence is maintained.

The first experiment to combine the concept of non-adiabatic alignment with HHG [12] has been the source of inspiration for many [13–16, 18]. While the notion of tomographic

imaging put forth in Ref. 12 remains controversial, it has been conclusively illustrated that harmonic signals from aligned systems provide useful information about the material system, which is not readily available from conventional spectroscopies. Thus, the two-center model of Ref. 14 shows that extrema in harmonic spectra provide information about the molecular internuclear distances - with femtosecond time-resolution in the case of transient structures.[13] Reference 15 illustrates that harmonic spectra from aligned rotational wave packets map the rotational coherences, hence providing information about dissipative mechanisms. The work of Refs. 16, 18 points to information regarding multiple orbital dynamics and ion-hole dynamics.

Particularly relevant to the effort to use HHG as a coherent spectroscopy, is a series of recent studies of the polarization of the emitted light.[19–21] For isotropic molecular media, symmetry considerations restrict the polarization of the generated harmonics to linear, but pre-aligned media introduce an opportunity to observe elliptically polarized harmonics, at least in principle. Provided that the angle ζ between the alignment and the ionization field vectors (see Fig. 1) differs from zero and $\pi/2$, the cylindrical symmetry is broken and the emitted light may be arbitrarily polarized. Although the original motivation of the search for ellipticity in HHG from pre-aligned media has been the interest in ultrafast birefringence in the XUV spectral range, it was soon realized that the ellipticity provides a sensitive probe of the material system. In particular, Ref. 25 illustrates that the ellipticity of the emitted light conveys information about the scattering phase of the continuum electronic wavefunction. That scattering phase is directly related to the molecular scattering potential.

Although symmetry in general allows the generation of elliptically polarized harmonics from aligned molecules, the question whether small linear molecules can in fact emit such light when driven by a linearly polarized source was a matter of controversy for several years. Whereas the experiments of Refs. 20, 22 found no ellipticity for either N_2 , O_2 or CO_2 molecules, the work of Ref. 23 detected experimentally considerable ellipticity from N_2 , over a wide range of harmonic orders. The numerical work of Ref. 20, carried out within the strong field approximation [5], found no ellipticity in the harmonics generated from N_2 molecules, in agreement with the measurements of Refs. 20, 22, whereas the theory of 24 suggested that preparation of the molecule in a state that carries electronic current may be necessary in order to observe ellipticity. The more recent study of Ref. 25 illustrated, both theoretically and numerically, significant ellipticity in the emission of N_2 molecules,

supporting the observations of Ref. 23. Furthermore, gratifying agreement between the numerical results and the experimental observations was demonstrated.

Several very recent numerical studies support the conclusions of Ref. 25. In particular, Ref. 26 shows that modified strong-field calculations taking into account trajectories from one atomic center to the other, can naturally account for ellipticity. Reference 27 illustrates quantitative agreement with the experimental observations of 23 and the calculations of 25 and 27 within the rescattering theory of Ref. 28.

The goal of the present study is threefold. First, we attempt to explore the relation of the observable ellipticity to the details of the molecular potential. To that end we adopt a very simple and commonly used model potential, where the availability of two parameters allows introducing careful modifications in the potential. Second, we provide the details of our theory, which were omitted from our earlier report[25] for space considerations. Last, we consider the sense in which, and degree to which, phase-sensitive observables, such as the ellipticity, are more sensitive to the system details and to the quality of the calculation than observables that do not contain phase information, such as the harmonic intensity. Our model has no ambition to describe quantitatively a specific molecule; rather, it is our intention to draw general qualitative conclusions. The model parameters, however, are chosen to correspond to the N_2 molecule.

In Sec. II, the theory and numerical scheme are presented, outlining the electronic dynamics and the angular momentum formulation. Section III presents and discusses the results and Sec. IV summarizes our conclusions. Atomic units are used throughout the paper, $\hbar = e = m_e = a_0 = 1$.

II. THEORY

We consider a molecular gas subject to two sequential laser pulses. The first (pump) pulse, ϵ_{pu} , a far-off-resonance pulse of duration short with respect to the rotational period and intensity below the ionization threshold, serves to align the molecule. The second (probe) pulse, ϵ_{pr} , a far-off-resonance intense pulse, is responsible for generation of high harmonics. Our coordinate system, depicted in Fig. 1, defines the polarization vector of the ionization pulse as the space-fixed z-axis, with the alignment field vector lying in the y-z plane. The angle between the polarization vectors is denoted ζ , $\zeta = \cos^{-1}(\hat{\epsilon}_{pu} \cdot \hat{\epsilon}_{pr})$, while

η denotes the angle between the z-axis and the polarization vector of the emitted light, $\eta = \cos^{-1}(\hat{n} \cdot \hat{\epsilon}_{pr})$, where $\hat{\epsilon}_{pu}$ ($\hat{\epsilon}_{pr}$) is a unit vector along the pump (probe) field polarization direction and $\hat{n}$ is a unit vector along the harmonics polarization vector. We follow the standard convention of denoting by θ and ϕ , respectively, the polar and azimuthal angles defining the orientation of the body-fixed with respect to the space-fixed frame.

The harmonic signal at frequency Ω is obtained as $I^\eta(\Omega) = |\mathbf{a}(\Omega) \cdot \hat{n}|^2$ where,

$$\mathbf{a}(\Omega) \cdot \hat{n} \propto \int \langle \Psi(t) | \mathbf{a}(t) \cdot \hat{n} | \Psi(t) \rangle e^{-i\Omega t} dt, \quad (1)$$

$\mathbf{a}(t)$ is the acceleration dipole, and $\Psi(t)$ is the complete electronic-rotational wavepacket. References 15, 29 show that by neglecting the rotational level spacing in comparison with the ionization potential (i.e., assuming that the rotations are slow with respect to the electrons) and taking advantage of the closure relation of the spherical harmonics, Eq. (1) can be cast in the form,

$$\langle \Psi(t) | \mathbf{a}(t) \cdot \hat{n} | \Psi(t) \rangle_T = \int d\hat{R} \rho_T^\zeta(\hat{R}; \tau) F_{\text{elec}}(R; t), \quad (2)$$

where the $F_{\text{elec}}(\hat{R}, t)$ contain the electronic dependence, with $\hat{R}$ denoting collectively the Euler angles of rotation. The rotational density matrix at temperature T , $\rho_T^\zeta(\hat{R}; \tau)$, is determined through solution of the rotational dynamics subject to the alignment pulse,[30]

$$\rho_T^\zeta(R; \tau) = \sum C_{JM}^\zeta(\tau) C_{J'M'}^{\zeta*}(\tau) Y_{JM}(\hat{R}) Y_{J'M'}^*(\hat{R}), \quad (3)$$

where J is the total angular momentum, M is its projection onto the space-fixed z-axis, τ is the pump-probe delay time, the Y_{JM} are spherical harmonics and the $C_{JM}^\zeta(\tau)$ are the associated expansion coefficients, containing the information about the pump pulse parameters and the alignment dynamics. The factors F_{elec} in Eq. (2) are given in terms of the electronic portion of the wave packet, $\psi(t)$, as,

$$F_{\text{elec}}(\hat{R}; t) = \langle \psi(t) | (\nabla V + \epsilon_{pr}(t)) \cdot \hat{n} | \psi(t) \rangle, \quad (4)$$

where V is the bound-state potential and $\epsilon_{pr}(t) = \hat{\epsilon}_{pr} \epsilon_{pr}(t)$ is the probe field.

The time-dependent Schrödinger equation for $\psi(t)$ is solved using the split-operator method[31] on a discrete Cartesian grid within the single active electron approximation. In the velocity gauge, chosen so to minimize nonphysical effects at the grid boundaries,[6] the electronic Hamiltonian is expressed as,

$$\hat{H}(t) = -\frac{1}{2} \nabla^2 - i\mathbf{A}(t) \cdot \nabla + V(\mathbf{r}), \quad (5)$$

where $\mathbf{A}(t) = \int \mathbf{E}(t')dt'$ is the vector potential of the driving field. The spatial domain required to accurately model the electron dynamics during the HHG process is large (10^7 a.u.^3), hence for computational tractability, we limit the Cartesian grid to two-dimensions, with which the electron motion is confined to the y-z plane (defined, Fig. 1, by the ionization and the alignment field polarization vectors). As discussed below, however, partial wave expansion of the electronic wavefunction and analysis of the dominating lowest terms allow us to approximate the neglected coordinate analytically.

As a simple analytic model potential we use the shielded soft-core potential, which has been often used successfully in prior studies[32]. In the case of a homonuclear diatomics it is written as,

$$V(\mathbf{r}) = - \sum_{i=1}^2 \frac{(Z^{\text{in}} - Z^{\text{out}}) \exp(-(r_i - r_e)^2/s^2)}{\sqrt{(r_i - r_e)^2 + \delta}}, \quad (6)$$

where Z^{out} and Z^{in} are the inner and outer effective charges per nucleus seen by the single electron, r_e is the equilibrium bond length, δ is a smoothing parameter and s describes the shielding of the nucleus by inner electrons. For the nitrogen molecule, $r_e = 1.0977 \text{ \AA}$, $Z^{\text{in}} = 5 \text{ a.u.}$ if the 1s electron region is considered as an effective core, and $Z^{\text{out}} = 0.5 \text{ a.u.}$, corresponding to a single charge shared by two nuclei. The two parameters s and δ are adjusted to accurately match the ionization potential of the molecule at hand. For the nitrogen molecule $I_p = 15.6 \text{ eV}$.

The degree of freedom neglected in the electronic dynamics can be taken into approximate account by formally expanding the continuum electronic function in partial waves, following Refs. 15, 29, and making use of symmetry. As sketched in what follows, this procedure suggests that, to low order in the partial wave expansion, the dependence of the F_{elec} on the azimuthal angle ϕ (for the specific case of N_2) is simple.

Expressing the electronic matrix elements in the body-fixed frame and performing partial-wave decomposition of the continuum electronic wavefunction, we have[15, 29]

$$\begin{aligned} \langle \Psi(t) | \mathbf{a}(t) \cdot \hat{n} | \Psi(t) \rangle = \int d\hat{R} \rho_T^\zeta(\hat{R}; \tau) \left\{ a_{\hat{n}}(\hat{R}) \sum_{l'l'k_lk_l'} Y_{lk_l}(\hat{R}) Y_{l'k_l'}^*(\hat{R}) F_{||}(l, l', k_l, k_{l'}, t) \right. \\ \left. + b_{\hat{n}}(\hat{R}) \sum_{l'l'k_lk_l'} Y_{lk_l}(\hat{R}) Y_{l'k_l'}^*(\hat{R}) F_{\perp}(l, l', k_l, k_{l'}, t) \right\}, \quad (7) \end{aligned}$$

where l is the electronic angular momentum, k_l denotes the projection of l onto the molecular axis, and the components of the electronic factors parallel and perpendicular to the

molecular axis are denoted $F_{\parallel}$ and $F_{\perp}$, respectively. The factors $a_{\hat{n}}(\hat{R})$ and $b_{\hat{n}}(\hat{R})$ are geometric functions of the rotation angles, deriving from the dot product $\mathbf{a}(t) \cdot \hat{n}$, whereas the electronic dynamics is entirely contained in the $F_{\parallel(\perp)}$.

It follows from the symmetry of the N_2 HOMO, that the even partial wave components of Eq. (7) (l even) vanish in both the parallel and perpendicular terms. Selection rules derived from the integrals in $F_{\parallel(\perp)}$ (see, for example, Ref. 33) also dictate that $k_l = k_{l'} = 0$ for the parallel case and $k_l = k_{l'} = \pm 1$ for the perpendicular. Furthermore, Ref. 15 shows that the partial wave expansion is rapidly convergent. Expanding the first series in equation (7) and neglecting terms higher than the third, one finds a ϕ -independent function,

$$\sum_{l'l'k_lk_{l'}} Y_{lk_l}(\hat{R}) Y_{l'k_{l'}}^*(\hat{R}) F_{\parallel}(l, l', k_l, k_{l'}, t) \approx \cos^2 \theta F_{\parallel}(1, 1, 0, 0, t) + (5 \cos^2 \theta - 3 \cos \theta) F_{\parallel}(3, 1, 0, 0, t). \quad (8)$$

The second (perpendicular) term can be shown to be independent of ϕ in a similar fashion but with terms involving $k_l = k_{l'} = \pm 1$ (terms where $k_l \neq k_{l'}$ cancel). Thus, the only significant dependence on ϕ results from the geometric factors determined through the dot products $\mathbf{a}(t) \cdot \hat{z}$ and $\mathbf{a}(t) \cdot \hat{y}$ and expressed through the $a_{\hat{n}}$ and $b_{\hat{n}}$ in Eq. (7). These can be readily shown to be,

$$a_z(\hat{R}) = \cos^2 \theta \quad b_z(\hat{R}) = \sin^2 \theta \quad (9)$$

for the z-component and

$$a_y(\hat{R}) = \sin \phi \cos \theta \sin \theta \quad b_y(\hat{R}) = -\sin \phi \cos \theta \sin \theta \quad (10)$$

for the y-components. With Eqs. (7,9) and (10), the dipole element in the directions parallel and perpendicular to the driving field are given as,

$$\begin{aligned} \langle \Psi(t) | \mathbf{a}(\Omega) \cdot \hat{z} | \Psi(t) \rangle &= 2\pi \int \sin \theta d\theta \rho_T^{\zeta}(\hat{R}; \tau) F_z(\theta, \Omega) \\ \langle \Psi(t) | \mathbf{a}(\Omega) \cdot \hat{y} | \Psi(t) \rangle &= \int \sin \theta d\theta d\phi \rho_T^{\zeta}(\hat{R}; \tau) \sin \phi F_y(\theta, \Omega) \end{aligned} \quad (11)$$

where

$$\begin{aligned} F_z(\theta) &= \left\{ \cos^2 \theta \sum_{l'l'k_lk_{l'}} Y_{lk_l}(\hat{R}) Y_{l'k_{l'}}^*(\hat{R}) F_{\parallel}(l, l', k_l, k_{l'}, t) \right. \\ &\quad \left. + \sin^2 \theta \sum_{l'l'k_lk_{l'}} Y_{lk_l}(\hat{R}) Y_{l'k_{l'}}^*(\hat{R}) F_{\perp}(l, l', k_l, k_{l'}, t), \right\} \end{aligned} \quad (12)$$

and

$$F_y(\theta) = \left\{ \cos \theta \sin \theta \sum_{l'l'k_lk'_l} Y_{lk_l}(\hat{R}) Y_{l'k'_l}^*(\hat{R}) F_{||}(l, l', k_l, k_{l'}, t) \right. \\ \left. + - \cos \theta \sin \theta \sum_{l'l'k_lk'_l} Y_{lk_l}(\hat{R}) Y_{l'k'_l}^*(\hat{R}) F_{\perp}(l, l', k_l, k_{l'}, t), \right\}. \quad (13)$$

In summary, the observed signal is simulated by solving separately for the slow (picosecond) rotational dynamics subject to the alignment pulse and the rapid (sub-femtosecond) electronic dynamics subject to the ionization pulse and applying Eq. (2). The former component is solved for exactly at a temperature T using the formulation developed in Ref. 15. The latter (electronic) component is approximated by performing two-dimensional electronic dynamics on a grid using a soft-core potential. Finally, the spectrum is obtained by Fourier transforming the acceleration dipole, as determined via Eq. (11)

III. RESULTS AND DISCUSSION

In this section, we apply the methodology discussed in Sec. II to the model potential defined by Eq. (6). The alignment (pump) pulse has a Gaussian envelope with a FWHM of 25 femtoseconds and peak intensity $7.5 \times 10^{13} \text{ Wcm}^{-2}$. The driving (probe) pulse peak intensity ($2 \times 10^{14} \text{ Wcm}^{-2}$) and frequency are chosen to correspond to typical experiments. The pulse envelope is a trapezoidal pulse of duration 10 optical cycles with 3 cycle ramps. The highest occupied molecular orbital is obtained by repeated imaginary time propagation,[34] at each step filtering out prior states to obtain a final state with the correct symmetry and the ionization energy of the $3\sigma_g$ state. The shielding and softening parameters are varied to change the potential function (see Fig. 2 and table I) and hence the orbital, whilst keeping the ionization potential and the bond length fixed at their physical values. A large grid of at least 512×128 a.u. is used, with spatial spacing 0.25 a.u. and $\cos^{2/3}$ mask functions that decay to zero at the grid boundaries. The mask functions are chosen to cover a length of 1/10 of the grid in the z direction and 1/8 of the grid in the y direction at each end. The masks are applied at each time step, and we use a step size that corresponds to 3072 steps per laser cycle ($\Delta t = 0.035$ a.u. for $\lambda = 800$ nm). The Fourier transform in Eq. (1) is performed under a Welch (quadratic) window function to minimize unwanted noise.

Softening parameter (δ , a.u.)	Shielding parameter (s , a.u.)
0.25	0.6958
0.35	0.7437
0.45	0.7844
0.50	0.8028

TABLE I: Softening parameter and corresponding shielding parameter for the ionization energy of nitrogen.

Ellipticity can be quantified in terms of the ratio of the major to the minor axis of the polarization ellipse and its angle of rotation with respect to the driving field polarization vector (the space-fixed z-axis). Related quantities that are experimentally observable are the intensity ratio between the emitted fields in directions parallel and perpendicular to the driving field and their phase difference, both are required to be non-zero for the harmonics to be elliptically polarized. In what follows we focus on these two observables.

The phase difference $\Delta\Phi$ was calculated at $T = 0K$ for a pump-probe time delay τ of a full rotational revival over a range of harmonic orders, 13 – 29, and a range of values of the softening parameter, $\delta = 0.25 - 0.5$ a.u. Two wavelengths were explored, $\lambda = 800\text{nm}$ - the standard choice of experiments, and a slightly detuned value, $\lambda = 780\text{nm}$, considered in order to unravel the physical origin of the structure of $\Delta\Phi$. In all cases, a significant phase difference is found, Fig. 3, pointing to significant ellipticity, irrespective of the model parameters. The phase difference is found to increase overall with harmonic order, as observed in experiments,[23] in our earlier study, where a much more accurate potential was used[25] and in the recent studies of Refs. 27 and 26. The disparity between the experiment and the simulation is expected, as the potential is simple and not intended to mimic reality. The oscillations observed in the 800 nm trace in Fig. 3 are an artifact of the soft-core potential and were not found in the studies of Refs. 23, 25–27. It is interesting to note, however, that they are stable with respect to variations in the model parameters and the parameters of the calculation. Separate calculations trace the origin of the structure in part to the presence of lower energy states in the model calculation - states of different symmetry from the highest occupied molecular orbital (HOMO) of the physical N_2 molecule, which lie below the proper symmetry state for the soft core Hamiltonian. More relevant to

the focus of the present study is the sensitivity of the phase difference to the parameters of the model potential. Interestingly, the harmonic intensities are invariant to the potential parameters over the same range, see Fig. 4. This remarkable result has been anticipated, since observables that are sensitive to the phase of the underlying continuum wavefunction (such as the ellipticity) carry much more information regarding the potential from ones that are not.

A different view of the physical picture described by Fig. 3 is presented in Fig. 5, which quantifies the response of the observable phase to the details of the potential. We find phase variations of order $0.03 - 0.16\pi$, depending on the particular harmonic. This sensitivity substantiates the suggestion[25] that the molecular potential is the main origin of the observed ellipticity. It is noted that in this work, as well as in our previous work[25] and in the work of 27, the phase difference is generally slightly lower than that found in experiment, which suggests that another mechanism may be involved.

Figure 6 provides the ratio between the y- and z-field components of the harmonics as a function of the harmonic order, over the range considered in Fig. 3 (see Fig. 1 for definition of our coordinate system). We note that the plot is normalized to a maximum ratio of unity. The perpendicular (y-) contribution to the harmonics is nonphysically large as compared to the parallel (z-) component, due to the reduced dimensionality of the electronic part of the simulation. On a qualitative level, this artifact is readily understood: the parallel component of the harmonics arises from driven electronic oscillations along the z-coordinate whereas the perpendicular component arises from oscillations in the xy-plane. Consequently, the perpendicular component is artificially magnified by the reduction of the dynamics to 2D, whereas the parallel component is not. Although the absolute magnitude of the ratio is exaggerated, trends can be gleaned. In particular, the scaled ratio is a strong function of the softening parameter δ , depending in sensitivity on the harmonic probed. Figure 7 summarizes the data of Fig. 6, illustrating variations ranging from an order of magnitude to a few percent, depending on the harmonic order. The results of Fig. 7 may have been anticipated. A softer potential has larger magnitude far from the nucleus, and hence has more influence on the phase of the continuum wavefunction. This effect is directly imprinted onto the polarization of the observed harmonics.

To further explore the generality of the ellipticity, we repeated the calculation for a set of parameters adjusted to correspond to the O_2 molecule. Here too we found a non-

negligible phase difference between the parallel and perpendicular components of the emitted harmonics over a wide range of harmonic orders.

The observation of a significant phase difference, and hence significant ellipticity, within a very simple and general model potential over a wide range of model parameters suggests that elliptically polarized harmonics from linear molecules driven by a linearly polarized field would be a rather general phenomenon, which, at the qualitative level, is primarily determined by the molecular symmetry and the properties of the bound state. The presence of ellipticity in the present calculation, as well as in the earlier studies that accounted for the molecular potential,[25–27] and its absence in strong field calculations give credence to the conclusion that the potential is at least part of the origin of elliptical emission under a linear driving field. Our findings support the conclusions of Ref. 25, where it is demonstrated analytically that the l -dependence of the phase of the partial wave scattering states is responsible for the observed phase difference. They are also consistent with the accurate calculations of Ref. 27 and with the recent study of Ref. 26, which proposes that ellipticity arises from recombination at a different nucleus than that from which ionization took place, requiring trajectories with angular momentum. The sensitivity of the magnitude of the phase difference and the amplitude ratio to the parameters of the Hamiltonian point to the potential of ellipticity measurements as a probe of the molecular potential, consistent with the conclusions of Ref. 23.

IV. SUMMARY AND CONCLUSION

We combined a previously developed formalism of high harmonic generation from nonadiabatically aligned molecules with a numerical approach for solving for the electronic dynamics quantum mechanically and an exact solution of the rotational dynamics, to explore the way in which, and degree to which, details of the molecular potential are imprinted onto the polarization of the emitted harmonics. To that end we used a simple and general model for the potential and systematically varied its parameters while keeping the ionization potential fixed. We considered specifically the case of N_2 molecules but several of our conclusions are more general. We found substantial ellipticity in the harmonics emitted from N_2 molecules subject to linearly polarized driving fields irrespective of the model parameters. The two observables characterizing the elliptical polarization of the emitted light, however,

vary markedly with variations in the potential.

Our findings are consistent with the results of Ref. 25, where it is demonstrated that properties of the phase of the partial wave scattering states is responsible for the observed ellipticity in recent experiments. They agree also with two more recent numerical studies[26, 27] that found ellipticity in the harmonic spectra of N_2 molecules. Finally, the above results point to the information content of harmonic signals regarding the electronic continuum wavefunction.

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Figure Captions

FIG. 1. Schematic illustration of the simulation setup.

FIG. 2. Slice through the molecular axis ($y = 0$) of the soft-core potential and its variation with the softening parameter. $\delta = 0.25$ (solid, black), $\delta = 0.35$ (dashed, red), $\delta = 0.45$ (dot-dashed, green), $\delta = 0.50$ (dotted, blue) .

FIG. 3. Phase difference in units of π as a function of the harmonic order for different driving field wavelengths, $\lambda = 780$ nm (black), $\lambda = 800$ nm (red), and softening parameters $\delta = 0.25$ (dotted), 0.35 (dot-dashed), 0.45 (dashed), 0.50 (solid).

FIG. 4. Harmonic spectrum at pump-probe angle $\frac{\pi}{4}$ radians, for softening parameters $\delta = 0.25$ (blue), 0.35 (green), 0.45 (red), 0.50 (black).

FIG. 5. Phase difference for a $\lambda = 800$ nm driving field as a function of the softening parameter for various harmonics. H13 (circles, black), H15 (squares, red), H17 (diamonds, green), H19 (triangles up, blue), H21 (triangles left, yellow), H23 (triangles down, brown), H25 (triangles right, grey), H27 (stars, violet), H29 (crosses, cyan).

FIG. 6. Ratio of the perpendicular to the parallel component of the emitted field as a function of harmonic order for different driving field wavelengths, $\lambda = 780$ nm (black), $\lambda = 800$ nm (red) and softening parameters $\delta = 0.25$ (dotted), 0.35 (dot-dashed), 0.45 (dashed), 0.50 (solid). The ratio is scaled to a maximum value of unity.

FIG. 7. Ratio of the perpendicular to the parallel component of the emitted field for a $\lambda = 800$ nm driving field as a function of the softening parameter for various harmonics: H19 (circles, black), H21 (squares, red), H23 (diamonds, green), H25 (triangles up, blue), H27 (triangles down, magenta), H29 (crosses, brown). Each harmonic is scaled to the maximum value at $\delta = 0.5$. Harmonics 13–17 are omitted for clarity.

Figures

FIG. 1:

FIG. 2:

FIG. 3:

FIG. 4:

FIG. 5:

FIG. 6:

FIG. 7: