## **Ellipticity Dependence of High-Order Harmonic Generation from Aligned Molecules**

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We report ellipticity dependence of high-order harmonic generation (HHG) from aligned N<sub>2</sub>, O<sub>2</sub>, and CO<sub>2</sub> molecules. Experimentally, we find that the ellipticity dependence is sensitive to molecular alignment and to the shape and symmetry of the valence orbitals. It is also found that the destructive interference in the recombination process affects the ellipticity dependence. Theoretically, we extend the original Lewenstein model to a more generalized model, which can be applicable to HHG from molecules, by introducing an electron acceleration parameter  $\xi_{\theta}$  and by combining the molecular orbital method. The present observations are successfully explained by our model.

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The high-order harmonic generation (HHG) from atoms and molecules has been a subject of intense studies for its applications as a coherent ultrashort radiation source in the extreme ultraviolet (XUV) and soft x-ray regions [1]. Recently, in addition to this motivation, Itatani *et al.* (with HHG [2]) and the present authors (with HHG and ionization [3]) proposed novel methods to take tomographic images of molecular structure with subfemtosecond time resolution using underlying physics of HHG, and this potential application has begun to attract much attention [4–6].

The basic physics of HHG is well understood by the 3step model [7]: First, a part of the bound-state electron wave function tunnels through the potential barrier modified by the intense laser field, and appears in the continuum (step 1). The freed electron wave packet is then driven by the laser field and after the field reverses its direction, it has a probability of returning to the molecule *without being accelerated by the potential of the parent ion* (step 2). The high-energy photon is emitted by the coherent oscillation between the returning electron wave packet and the boundstate electron wave function (step 3).

The 3-step model tells us that harmonics are generated only when the freed electron returns to the ionic core. When the ellipticity  $\epsilon$  of the laser pulse is increased from zero, the electron is driven away by the transverse field component from its parent ion so that the harmonic intensity falls off rapidly [8]. This strong dependence of HHG on the ellipticity is employed to generate single attosecond pulses with the polarization gating method [9].

As long as a sample of randomly aligned molecules is employed, HHG from molecules have shown characteristics similar to those from atoms [10]. This holds true also for ellipticity dependence of HHG [11,12], though Shan *et al.* observed the difference in the ellipticity dependence between randomly aligned N<sub>2</sub> and O<sub>2</sub> molecules [12]. In order to observe alignment effects in detail, we need to prepare a sample of aligned molecules. Based on recent developments of molecular alignment [13–15] and orientation [16] techniques, HHG from molecules has been found to be sensitive to the molecular alignment [2,3,17].

When we adopt aligned molecules as a nonlinear medium, it is natural to expect that the ellipticity dependence should depend on the molecular alignment. Since recent studies with linearly polarized pulses indicate that the HHG from molecules reflects the structure of valence orbitals [2,3,17], revealing ellipticity dependence of HHG from aligned molecules provides further deep insight into the underlying physics of HHG, i.e., the basis of the ultrafast imaging of molecular structures.

In this Letter, we demonstrate that the dependence of harmonic intensity from *aligned*  $N_2$ ,  $O_2$ , and  $CO_2$  molecules on the ellipticity of the driving laser field is sensitive to the molecular alignment and the structures of valence orbitals. In addition, we report that the destructive interference (DI) [3] also appears in the ellipticity dependence. We have developed an analytical theory of HHG for molecules by modifying the Lewenstein model using molecular orbitals with correct asymptotic behavior, and we succeeded in explaining all of the experimental results.

An output from a Ti:sapphire based chirped pulse amplification system with a pulse width of  $\sim 50$  fs and a center wavelength of  $\sim 800$  nm is split into two pulses. The first linearly polarized pulse is used as a pump to create rotational wave packets and to induce nonadiabatic molecular alignment. The second pulse with elliptical polarization is delayed by a computer controlled translation stage and is used as a probe to generate harmonics. The ellipticity of the second pulse is adjusted by rotating a zero order half-wave plate placed before a fixed zero order quarter-wave plate. Thereby, one can use an arbitrary ellipticity with the directions of the two axes of the ellipse fixed in the laboratory frame. The intensity of the pump pulse is  $\sim 6 \times 10^{13}$  W/cm<sup>2</sup> and that of the probe is  $\sim 2 \times$  $10^{14}$  W/cm<sup>2</sup>, which is well below the saturation intensity of molecular ionization. The generated harmonics are spectrally resolved by a 1 m grazing incidence monochromator and detected by an electron multiplier.



FIG. 1 (color online). (a) An illustration of the two configurations used in the experiment. (b) The ellipticity dependence of the 25th harmonic intensity from aligned CO<sub>2</sub> molecules in the parallel (blue squares) and perpendicular (red circles) cases as a function of ellipticity  $\epsilon$  of the fundamental pulse. Harmonic intensities are normalized to those at  $\epsilon = 0$  (linear polarization). The error bars are about the sizes of the symbols for the experimental data. The results of theoretical calculations are shown by a blue curve for the parallel case and a red curve for the perpendicular case, respectively.  $\Delta \epsilon_{\parallel}(CO_2) \sim 0.20$ , and  $\Delta \epsilon_{\perp}(CO_2) \sim 0.26$ . See text for the definition of  $\Delta \epsilon$ .

Based on the relation between molecular axis and major axis of the ellipse of the laser field, here we use two configurations depicted in Fig. 1(a), i.e., the perpendicular and parallel cases. Figure 1(b) shows the ellipticity dependence of the 25th harmonics from aligned CO<sub>2</sub> molecules in the parallel (blue squares) and perpendicular (red circles) cases. The pump-probe delay is fixed at 20.9 ps, which corresponds to the delay of half revival when  $CO_2$ molecules are aligned [2,3,14]. As can be seen, the ellipticity dependence in the perpendicular case is different from that in the parallel case. In order to discuss the ellipticity dependence quantitatively, we introduce a parameter  $\Delta \epsilon$  which is defined as a range of ellipticity of the probe pulse that can generate harmonics at an efficiency of more than 20% of those generated by the pulses with linear polarization. Using this parameter, Fig. 1(b) shows that  $\Delta \epsilon_{\parallel}(\text{CO}_2) \sim 0.20$  and  $\Delta \epsilon_{\perp}(\text{CO}_2) \sim 0.26$  for CO<sub>2</sub>. That is, the  $\Delta \epsilon$  in the perpendicular case is 1.3 times as large as that in the parallel case. This relation of  $\Delta \epsilon_{\parallel} < \Delta \epsilon_{\perp}$  is also observed for the 25th harmonics from aligned O2 molecules whose valence orbitals have the same  $\pi_g$  symmetry  $[\Delta \epsilon_{\parallel}(O_2) \sim 0.22$  and  $\Delta \epsilon_{\perp}(O_2) \sim 0.28]$ . Roughly speaking, since the valence orbital ranges along the molecular axis, the electron can return to the core even with larger ellipticity in the perpendicular case.

This intuitive alignment dependence is also observed for molecules whose valence orbitals have  $\sigma_g$  symmetry. Figure 2(a) shows the ellipticity dependence of the 23rd harmonics from aligned N<sub>2</sub> molecules in the parallel (blue squares) and perpendicular (red circles) cases. The pumpprobe delay is fixed at 4.1 ps, which corresponds to the delay of half revival when N<sub>2</sub> molecules are aligned [2,3,14]. From the data shown in Fig. 2(a), we obtain  $\Delta \epsilon_{\parallel}(N_2) \sim 0.19$  and  $\Delta \epsilon_{\perp}(N_2) \sim 0.21$ . For the 23rd harmonics, the harmonic intensity from N<sub>2</sub> monotonically decreases as in the case of atoms like Ar when the ellipticity is increased. Comparing the results from O<sub>2</sub> and N<sub>2</sub>,



FIG. 2 (color online). (a) The same as in Fig. 1(b) but for the 23rd harmonic intensity from aligned N<sub>2</sub> molecules.  $\Delta \epsilon_{\parallel}(N_2) \sim 0.19$  and  $\Delta \epsilon_{\perp}(N_2) \sim 0.21$ . (b) The ellipticity dependence of the 31st harmonic intensity from N<sub>2</sub> molecules measured in the parallel case (blue squares) together with that from Ar (red circles). The error bars are about the sizes of the symbols for the experimental data.

we obtain  $\Delta \epsilon_{\parallel,\perp}(N_2) < \Delta \epsilon_{\parallel,\perp}(O_2)$ , which is consistent with the results by Shan *et al.*,  $\langle \Delta \epsilon(N_2) \rangle < \langle \Delta \epsilon(O_2) \rangle$  for the 45th harmonics [12].

To understand the experimental results shown in Figs. 1(b) and 2(a), we applied our analytical model [3] to the ellipticity dependence. The model is built by combining the Lewenstein model and the molecular orbital method. The numerical models are not necessarily robust especially for molecules with an antibonding orbital, since the orbital is not the ground state in the multiwell potential of the form,  $V(\mathbf{x}) = -\sum_i [1/\sqrt{(\mathbf{x} - \mathbf{R}_i)^2 + a}]$ , where  $\mathbf{R}_i$ 's are the positions of the nuclei [18,19]. One of the advantages of our model is that we can exclude the inappropriate states below the valence orbital from the basis set. We integrate the equation analytically rather than numerically.

Without using the inappropriate states, the Lewenstein model [20] combined with the molecular orbital method is used to calculate the harmonic spectrum from the *molecular* dipole moment in the time domain,

$$\mathbf{x}(t) = i \int_0^\infty d\tau \left(\frac{\pi}{\nu + i\tau/2}\right)^{3/2} \mathbf{D}^* [\mathbf{p}_{st} - \mathbf{A}(t)] e^{-iS(\mathbf{p}_{st}, t, \tau)} \\ \times \mathbf{E}(t - \tau) \cdot \mathbf{d} [\mathbf{p}_{st} - \mathbf{A}(t - \tau)] + \text{c.c.},$$
(1)

where  $\nu$  is a regularization constant,  $p_{st}(t, \tau) =$  $\int_{t-\tau}^{t} dt'' A(t'') / \tau$  is the canonical momentum corresponding to the stationary phase. In order to develop the most general theoretical model, here we consider elliptical polarization for the driving pulse, for which linear and circular polarizations are included as special cases. We take the x axis along the direction of the major component of the field and the y axis along the direction of the minor component, so that the vector potential and the electric field of the laser pulse are given by  $\boldsymbol{A}(t) = 2\sqrt{U_p/(1+\epsilon^2)}[\boldsymbol{e}_x\cos(t) + \boldsymbol{e}_y\epsilon\sin(t)],$  $\boldsymbol{E}(t) =$  $2\sqrt{U_p/(1+\epsilon^2)}[\boldsymbol{e}_x \sin(t) - \boldsymbol{e}_y \epsilon \cos(t)], \quad \text{respectively.}$   $S(\boldsymbol{p}, t, \tau) = \int_{t-\tau}^t dt'' \{[\boldsymbol{p} - \boldsymbol{A}(t'')]^2/2 + I_p\}, \text{ is the action}$ of the electron moving in the laser field.  $I_p$  is the ionization potential of the molecule.  $d(p) := \langle p | \hat{x} | 0 \rangle = (2\pi)^{-3/2} \times$  $\int_{-\infty}^{\infty} d^3 \mathbf{x} \exp(-i\mathbf{p} \cdot \mathbf{x}) \mathbf{x} \langle \mathbf{x} | 0 \rangle$  is the bound-free dipole transition matrix element between the ground state  $|0\rangle$  and the continuum state  $|\mathbf{p}\rangle$ .  $\mathbf{D}[\mathbf{p}] := d\{[\mathbf{p}^2 + 2(1 - \xi_{\theta})I_p]^{1/2}\hat{\mathbf{p}}\}$ is the modified bound-free transition dipole moment that we propose in order to include the effect of electron acceleration in the vicinity of the parent ion before recombination with the refraction effect neglected. The electron acceleration parameter  $\xi_{\theta}$  ( $0 \le \xi_{\theta} \le 1$ ) is defined so that  $\xi_{\theta} = 0$  corresponds to the case of  $I_p = 0$  [21] and  $\xi_{\theta} = 1$ corresponds to the case in which no acceleration takes place [2]. In general,  $\xi_{\theta}$  is a function of the orientation angle  $\theta := \arccos(\hat{p} \cdot \hat{R}) \ (0 \le \theta \le \pi)$ , where  $\hat{R}$  is a unit vector along molecular axis. Through recent experimental studies on HHG in molecules [2,3], we conclude that the Lewenstein model should be modified in the recombination process even for atomic cases. Although Lewenstein et al. calculated dipole moments only for center-symmetric atomic orbitals, our model enables us to deal with any valence orbital with different symmetries. We first calculate a complicated valence orbital in the configuration space  $\langle \mathbf{x} | 0 \rangle \equiv \Psi(\mathbf{x})$  using the molecular orbital method, and then calculate an induced time-dependent dipole moment using our analytical method.

For  $N_2$ ,  $O_2$ , and  $CO_2$  molecules whose valence orbitals have even parity symmetry, the valence orbital can be expressed by

$$\Psi_g(\mathbf{x}) = N_i [\Phi(\mathbf{x} + \mathbf{R}/2) + \Phi(-\mathbf{x} + \mathbf{R}/2)]. \quad (2)$$

Here  $N_i$ 's are normalization factors,  $\Phi$  is the atomic orbital,  $\mathbf{R}/2$  and  $-\mathbf{R}/2$  are the positions of the nuclei (for CO<sub>2</sub> molecules, the positions of the two O atoms [3]). The equilibrium internuclear separation  $|\mathbf{R}| \equiv R$  is 0.110 nm for N<sub>2</sub>, 0.121 nm for O<sub>2</sub>, and 0.232 nm for CO<sub>2</sub>.

For O<sub>2</sub> and CO<sub>2</sub> with the valence orbital of  $1\pi_g$ , since  $\Phi$  can be expressed by pure 2p orbitals, the valence orbital is simply given by

$$\Psi_{1\pi_g}(\mathbf{x}) = N'[\Phi_{2p_{y'}}(\mathbf{x} + \mathbf{R}/2) - \Phi_{2p_{y'}}(\mathbf{x} - \mathbf{R}/2)], \quad (3)$$

where  $\Phi_{2p_{y'}}(\mathbf{x})$  is the atomic 2*p* orbital in the configuration space that is aligned perpendicular to the molecular axis. Here we adopt a Slater-type basis, which has the correct asymptotic behavior ~  $\exp(-\alpha |\mathbf{x}|)$  as  $|\mathbf{x}| \rightarrow \infty$ , rather than a Gaussian-type basis (~  $\exp(-\alpha \mathbf{x}^2)$ ), which falls off more rapidly than a Slater-type basis does. A Gaussiantype basis can lead to a large error in the calculation since the bound-free transition is largely determined by the asymptotic tail of the ground state wave function [20,22].

Then the dipole transition moments for  $O_2$  and  $CO_2$  are given by

$$d_{1\pi_g}(\boldsymbol{p}) = N_{1\pi_g} [2i\sin(\boldsymbol{p} \cdot \boldsymbol{R}/2)d_{2p_{y'}}(\boldsymbol{p}) -\cos(\boldsymbol{p} \cdot \boldsymbol{R}/2)\tilde{\Phi}_{2p_{y'}}(\boldsymbol{p})\boldsymbol{R}], \qquad (4)$$

where  $d_{2p_{y'}}(p)$  is the atomic dipole moment from the  $2p_{y'}$  orbital, and  $\tilde{\Phi}_{2p_{y'}}(p)$  is the  $2p_{y'}$  wave function in the momentum space.

The first term in Eq. (4) is the product of two parts and its physical meaning can be understood by the simple twopoint emitter model [21]. The latter part  $[d_{2p_y}(p)]$  is the atomic counterpart and the former part  $[\sin(p \cdot R/2)]$  is the interference between the two atomic wave functions in the configuration space. Notice that this term is zero when

$$\boldsymbol{p} \cdot \boldsymbol{R}/2 = n\pi \Leftrightarrow R\cos\theta = n\lambda, \tag{5}$$

where *n* is an integer, *p* and  $\lambda = 2\pi/|p|$  are the momentum and the wavelength of the recombining electron, respectively, and  $\theta$  is the angle between *p* and *R*. The right-hand side of Eq. (5) is the condition for DI, that can be derived from the simple two-point emitter model [21]. Similarly,  $p \cdot R/2 = (n - 1/2)\pi$  leads to the condition for constructive interference. In Eq. (5), a special case of  $\theta = \pi/2$  is met by taking n = 0 and the physics can be understood by the intuitive semiclassical model [18].

The second term in Eq. (4) [23] corrects an error of the simple two-point emitter model [12,24], which has not been pointed out in Ref. [25]. This term is oriented along the molecular axis. Since  $\cos^2 + \sin^2 = 1$ , the magnitude of the first term and that of the second term anticorrelate to each other. This means that, if we use molecules with large R, we can control the polarization of harmonics via DI.

For N<sub>2</sub> molecules, on the other hand, it is difficult to give a simple explanation for the HHG process, because the valence orbital  $3\sigma_g$  of N<sub>2</sub> has a complicated structure [6]. N<sub>2</sub>  $3\sigma_g$  is constructed not only by atomic 2*p* orbitals but by 1*s* and 2*s* orbitals about 30%, whose symmetry is different from that of 2*p* orbitals. This complicated structure of N<sub>2</sub> makes the simple two-center interference picture problematic. In fact, different interference patterns are expected for the two types of orbital. While the two *s* orbitals are added in phase and the condition for constructive interference is given by Eq. (5), the *p* orbitals are added in opposite phase, leading to interchanged conditions for constructive and destructive interferences.

It should be noted that the molecules are not perfectly aligned and have angular distributions in both parallel and perpendicular cases. The observed high-order harmonic signals in Figs. 1(b) and 2(a) are the superpositions of the radiation from all the molecules in the interaction region. The ellipticity dependence of harmonic intensity,  $\langle I(\Omega)\rangle(\epsilon)$ , is given by  $\langle I(\Omega)\rangle(\epsilon) = |\int d\Omega \rho(\Omega) x(\Omega, \epsilon)|^2$ , where  $\rho(\Omega)$  is the angular distrubution of the molecules calculated by integrating the time-dependent Schrödinger equation of molecular rotation according to [3]. For simplicity, we assume that molecules with the same orientation angle make the same contribution to HHG. As can be seen in Figs. 1(b) and 2(a), we see satisfactory agreement between the experiments and the calculations, which ensures the validity of our model.

In our recent paper [3], we observed DI using  $CO_2$  molecules. The DI also appears in the ellipticity dependence of N<sub>2</sub>. Blue squares in Fig. 2(b) shows the measured ellipticity dependence of the 31st harmonics from aligned



FIG. 3 (color online). Theoretical calculations based on the molecular Lewenstein model. (Left) Harmonic intensity from N<sub>2</sub> molecules with linear polarization as a function of harmonic order and the orientation angle  $\theta$ . (Right) Intensity of the 31st harmonics from N<sub>2</sub> molecules as a function of ellipticity  $\epsilon$  and the orientation angle  $\theta$ . The laser parameters are as in Fig. 1 and  $\xi_{\theta} = 1$  is assumed.

 $N_2$  in the parallel case. For the purpose of comparison, the ellipticity dependence of the 31st harmonics from Ar (red circles), whose ionization potential is almost the same as that of  $N_2$ , is also plotted in Fig. 2(b). In the case of aligned  $N_2$ , we observe a suppression at ellipticity  $\epsilon \sim 0$  and a peak at  $\epsilon \sim 0.05$ . However, in the case of Ar, no suppression is observed at  $\epsilon \sim 0$ . The results observed for  $N_2$  can be understood as a corollary of the three-step model [7]. The generation of the 31st harmonics by pulses with linear polarization ( $\epsilon \sim 0$ ) is suppressed because of the DI. As the ellipticity of the driving laser field becomes larger, a transverse field component gives a transverse component of the returning electron momentum, making the DI less effective in the recombination process.

With our model, we calculated harmonic spectra from  $N_2$  molecules for various orientation angles (Fig. 3, left). Although, in general, N2 molecules aligned along the driving laser field (small  $\theta$ ) efficiently generate harmonics, we can see significant suppression of harmonic generation at around 30th harmonics which can be interpreted as the result of DI. Furthermore, we calculated the 31st harmonic intensity as a function of ellipticity  $\epsilon$  and orientation angle  $\theta$  (Fig. 3, right). In fact, the DI with linear polarization  $(\epsilon \sim 0)$  is expected in the ellipticity dependence at the orientation angle  $\theta \sim 0$ , which is consistent with our observation shown in Fig. 2(b). Our model shows that harmonic intensity from O<sub>2</sub> and CO<sub>2</sub> molecules can also be peaked at nonzero ellipticity depending on the electron acceleration parameter  $\xi_{\theta}$ . This effect offers the possibility of enhancing specific harmonics by varying the laser ellipticity and determining the value of  $\xi_{\theta}$  experimentally. Note that harmonic generation in an atom is always peaked at  $\epsilon = 0.$ 

In conclusion, we observed two distinct effects on ellipticity dependence of HHG from aligned molecules. One is an intuitive alignment effect, and the other is an effect from DI [3]. All of our observations were successfully explained by our new theoretical model. Using aligned molecules as a nonlinear medium for HHG can serve as a new route to control not only polarization of harmonics [Eq. (4)] but also the pulse width of harmonics with the potential for the generation of attosecond pulses via the polarization gate method [9].

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