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Separation of Target Structure and Medium Propagation Effects in High-Harmonic Generation

Cheng Jin,[1] Hans Jakob Wörner,[2] V. Tosa,[3] Julien B. Bertrand,[2] Anh-Thu
Le,[1] R. R. Lucchese,[4] P. B. Corkum,[2] D. M. Villeneuve,[2] and C. D. Lin[1]

[1] *J. R. Macdonald Laboratory, Physics Department,*
Kansas State University, Manhattan, Kansas 66506-2604, USA
[2] *Joint Laboratory for Attosecond Science, National Research Council of Canada and University of Ottawa,*
100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6
[3] *National Institute for R&D of Isotopic and Molecular Technologies, 400293 Cluj-Napoca, Romania*
[4] *Department of Chemistry, Texas A&M University, College Station, Texas 77843-3255, USA*
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We calculate high-harmonic generation (HHG) by intense infrared lasers in atoms and molecules with the inclusion of macroscopic propagation of the harmonics in the gas medium. We show that the observed experimental spectra can be accurately reproduced theoretically despite that HHG spectra are sensitive to the experimental conditions. We further demonstrate that the simulated (or experimental) HHG spectra can be factored out as a product of a "macroscopic wave packet" and the photo-recombination transition dipole moment where the former depends on the laser properties and the experimental conditions, while the latter is the property of the target only. The factorization makes it possible to extract target structure from experimental HHG spectra, and for ultrafast dynamic imaging of transient molecules.

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High-harmonic generation (HHG) has been employed to probe electronic structure of molecules on an ultra-fast time scale in recent years [1–3]. When molecules are placed in an intense laser field, electrons that are removed earlier may be driven back to recollide with the parent ion. HHG occurs when the returning electrons recombine with the parent ion with the emission of high-energy photons as in an inverse photoionization (PI) process. Since PI is a sensitive tool for probing electronic structure of molecules, HHG may serve likewise, but with the advantage of ultrafast temporal resolution as well as covering a coherent broad spectral range from XUV to soft-X-rays. Experimentally, however, HHG is generated from all the molecules in the interaction region. The radiations from them co-propagate with the fundamental infrared (IR) beam nonlinearly. To extract structure information of individual molecules, e.g., the amplitude and phase of PI transition dipole from the measured HHG, the propagation effect in the medium should be investigated. For molecular targets, this has not been done so far. Instead, it was often assumed that HHG was measured under the perfect phase-matching condition and that the observed harmonics were directly proportional to the harmonics from a single molecule. While such assumptions may be adequate for explaining many experimental observations qualitatively, such as the dependence of HHG on molecular alignment and on symmetry of the molecular orbital, the two-center interference [4, 5], and multiple-orbital contributions to HHG [6], they are inadequate if accurate structure information of individual molecules are to be extracted from the observed HHG spectra.

The effect of macroscopic propagation (MP) on the observed HHG spectra for atoms has been investigated extensively in the past two decades in connection with the generation of attosecond pulses. However, we are not aware of any direct comparison between experimental HHG and theoretical simulations over an extended spectral region. The Maxwell's equations that govern the propagation of the fundamental driving IR field and the generated harmonics are well established. To carry out such propagation calculations, accurate induced atomic dipoles generated by lasers for hundreds of peak intensities should be calculated which serve as the source term of the harmonics. These induced dipoles are often calculated using the strong-field approximation (SFA), or the so-called Lewenstein model [7]. The SFA does not describe the laser-atom interactions accurately, thus the results from the propagation can only be used to qualitatively interpret the experiments. While accurate induced dipoles can be obtained from solving the time-dependent Schrödinger equation (TDSE), the calculation is rather time consuming and was rarely attempted except for very few occasions [8]. Thus after two decades, our understanding of experimental HHG data is still mostly at the qualitative level.

In this Letter, we show that such limitations have been removed. We generate HHG spectra theoretically under experimental conditions and the results are compared directly to the observed data. The simulated spectra agree well with the measured one, over a broad photon energy region. The experiments were taken in a geometry as depicted in Fig. 1(a), using IR laser pulses with wavelengths of 1200 nm and 1360 nm, respectively, generated in a high-energy optical parametric amplifier (HE-TOPAS).

The TOPAS was pumped by the output of a multipass femtosecond amplifier system (8 mJ, 32 fs, 50Hz). High-harmonic radiation generated from the gas jet is allowed to propagate and a slit is placed downstream. After the slit, a concave grating disperses the harmonics which are then detected with a CCD detector. The gas jet is formed from a supersonic expansion of Ar or N_2 at a stagnation pressure of 3 bars. Experiments were performed on either isotropic or aligned N_2 molecules.

Fig. 1(b) shows the HHG spectra of Ar generated by a 1200 nm laser. The horizontal axis is the photon energy and the vertical axis is the transverse spatial dimension. The upper frame is from the measurement, while the bottom frame is from the simulation. There is a general agreement of the two spectra. The "up-down" asymmetry in the experimental HHG spectra is due to asymmetry in the laser beam profile. The faint features near 50 eV are the "famous" Cooper minimum in Ar [9], observed in photoionization, as well as in earlier HHG spectra [10, 11]. The harmonic yields integrated over the vertical dimension are compared in the upper half of Fig. 1(c). The lower half shows the HHG spectra taken with 1360 nm laser. In both cases, we can see very good agreement between theory and experiment over the 30-90 eV region covered. Experimentally, the gas jet is 0.5 mm long and placed 3 mm after the focus. A vertical slit with a diameter of 100 μm is placed 24 cm after the gas jet. For the 1200 (1360) nm the beam waist at the laser focus is 47.5 (52.5) μm, and the pulse duration is $\sim$40 ($\sim$50) fs. To achieve best overall agreement, in the simulation the peak intensity and gas pressure for each wavelength are adjusted till best overall fit in the data are achieved. Thus for 1200 nm laser, the peak intensity for experiment (theory) is 1.6 (1.5)$\times 10^{14}$W/cm^2, gas pressure is 28 (84) Torr. For the 1360 nm laser, the corresponding parameters are 1.25 (1.15)$\times 10^{14}$W/cm^2, and 28 (56) Torr, respectively.

In the theoretical simulation, we first obtain single-atom induced dipole using the quantitative rescattering (QRS) theory [12–14]. The Ar is treated in the single-active electron approximation using model potential proposed by Muller [15]. The resulting induced dipole for different peak intensities are then fed into the Maxwell's equations. The propagation equations for the fundamental and the harmonics are the standard ones [16–18]. For Ar target, we include dispersion, absorption, Kerr and plasma effects on the fundamental field in the medium. For the harmonics, only the dispersion and absorption are included. The harmonic yields emitted at the exit face of the gas jet (near field) are propagated to the far field where the harmonics are measured. They are obtained from the near-field harmonics through a Hankel transform [19, 20]. We assume the laser beam in the entrance of gas jet has the Gaussian shape.

A careful examination of Figs. 1(b) and (c) reveals that there are still small discrepancies between the experimen-
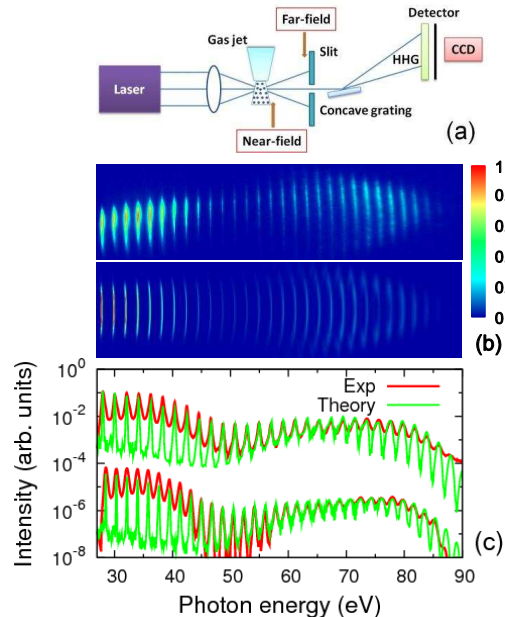

FIG. 1: (Color online) (a) Typical configuration for measuring HHG in the far field. (b) HHG spectra of Ar generated by 1200 nm laser. Upper frame: experiment; lower frame: theory. (c) Comparison of theoretical (green curves) and experimental (red curves) HHG yields integrated over the vertical dimension for 1200 nm (upper curves) and 1360 nm (lower curves) lasers. Laser parameters are given in the text.

tal data and the simulation. The harmonic width (or harmonic chirp) is narrower from the theory than from the experiment. Harmonic chirp is a direct consequence of temporal variation of laser intensity. The harmonic width is mainly influenced by the pulse duration, pressure, and laser intensity [21–23]. The width decreases with increasing pulse duration, and with increasing gas pressure. Other experimental factors like the use of slits and location of the detector also can affect the HHG spectra.

High-order harmonics from molecules by 1200 nm lasers have been reported for aligned and randomly distributed N_2 and CO_2 recently [24]. Here we report our simulated results for N_2, at the two peak laser intensities, 0.9 and 1.1$\times 10^{14}$W/cm^2, reported in [24]. To achieve good agreement in the cutoff positions, the two intensities used in the theory are 0.78 and 0.9$\times 10^{14}$W/cm^2 instead, respectively. Since the experiment was carried out at low laser intensity and low gas pressure, the harmonics are propagated without absorption and dispersion effects from the medium, and the fundamental laser field is not modified through the medium [25]. In the theoretical simulation, we first obtain induced dipoles of fixed-in-space molecules using QRS theory [13, 14] for different laser peak intensities. The induced dipoles are averaged coherently according to the alignment distribu-
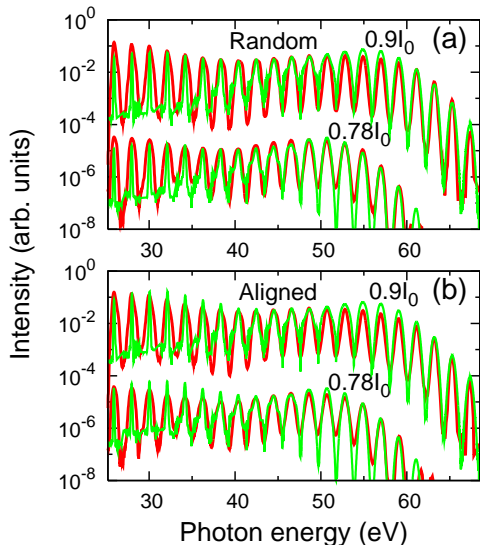
FIG. 2: (Color online) Comparison of HHG spectra from theory (green curves) and experimental results (red curves) of Ref. [24], (a) for randomly distributed N_2 and (b) for N_2 aligned along laser polarization direction. The laser intensities are indicated where $I_0 = 10^{14}$ W/cm^2. See text for additional laser parameters.

tion and then fed into the Maxwell's equations. Fig. 2 shows the good overall agreement between the measured and the simulated spectra, for both randomly distributed and aligned N_2. By examining the experimental HHG spectra more carefully, they reveal a shallow minimum at 38 ± 2eV (low intensity) and at 41 ± 2eV (high intensity) for both aligned and unaligned molecules. The theory also predicts a minimum: for unaligned molecules, the minimum is at $\sim$39eV for low intensity and $\sim$40eV for high intensity. For aligned molecules, the minimum is at $\sim$42eV for low intensity and $\sim$44eV for high intensity. In the experiment, the degree of alignment was estimated to be $\langle \cos^2 \theta \rangle = 0.6$-0.65. In the simulation, an alignment distribution of $\cos^4 \theta$ is used. Note that only HOMO is included in the calculation. We believe that this is the first time that HHG from molecules have been calculated including the propagation effect in the medium and the simulated results have been compared directly to the measured spectra. In the future, HHG spectra taken at different alignment angles should be compared together. Such comparison would help to identify factors that contribute to the remaining discrepancies between experiment and simulation.

The macroscopic HHG spectra can be expressed as [25]

$$S_h(\omega) \propto \omega^4 |W(\omega)|^2 |d(\omega)|^2 \qquad (1)$$

where $W(\omega)$ (the complex amplitude) is called "Macroscopic wave packet" (MWP), $d(\omega)$ is PI transition dipole moment for the atom. For the molecule $d(\omega)$ is taken to be the coherently averaged PI transition dipole mo-

ment $d^{avg}(\omega) = \int_0^\pi N(\theta)^{\frac{1}{2}} \rho(\theta) d(\theta, \omega) \sin \theta d\theta$, where $N(\theta)$ is the alignment-dependent ionization probability, $\rho(\theta)$ is the alignment distribution, and $d(\theta, \omega)$ is alignment-dependent transition dipole moment [26]. The polarization of pump laser is assumed parallel to probe laser. For unaligned molecules, $\rho(\theta)$ is a constant. Actually, MWP has the clear physical meaning. It can be considered as the collective effect of microscopic wave packets for the returning electrons [13, 14], which is governed by Maxwell's equations. In other words, the laser and macroscopic medium effects are all combined into MWP.

The validity of Eq. (1) has been checked in Jin et al. [25] when both the laser intensity and the gas pressure are low. The correctness of this relation has been assumed in Itatani et al. [1] by comparing Ar with N_2, and in Levesque et al. [27] for rare gas atoms. However, this relation has not been carefully checked for different focusing conditions and laser parameters. Theoretically, we have checked the validity of Eq. (1) carefully. We carried out macroscopic propagation calculation of HHG using single-atom (single-molecule) induced dipole obtained by QRS and by SFA. We have been able to show that the MWP obtained from the two calculations agree rather well, irrespective of laser parameters or the focusing conditions. In other words, the medium propagation only affects HHG through its modifications on the MWP. In this way, to study propagation effect on HHG, we can just study how the MWP (only the amplitude is considered below) depends on the lasers and the experimental conditions.

In Fig. 3(a) we show the dependence of MWP on the position of the Ar gas jet with respect to the laser focus. The laser intensity is 1.6×10^{14}W/cm^2, and the gas pressure is 56 Torr. For easy visualization we show the smooth envelope of $|W(\omega)|$. The three curves are for the gas jet at z=-3 (before), 0 (at), +3 (after) mm with respect to the laser focus. It is generally known that HHG achieves best phase matching if the gas jet is placed behind the laser focus where the dipole phase from the harmonic can be partially canceled by the Gouy phase. Thus among the three curves, the "after" curve is the flattest one. For the "before" focus, the MWP varies most as the photon energy is changed, and in the HHG spectra, the width of each harmonic (not shown) is not sharp, and the phase varies widely from order to order — reflecting the poor phase matching for this geometry.

In Fig. 3(b) we compare the MWP derived from changing the Ar gas pressure for the "after" focusing condition. The MWP has been normalized by the ratio of the pressure. Under perfect phase matching condition, the MWP $|W(\omega)|$ (the amplitude) should be proportional to the pressure [28]. The three curves are on top of each other from 45-75 eV, indicating good phase-matching in this energy region, but differ somewhat at lower and higher energies, indicating good phase matching condition is not fulfilled. This demonstrates that phase matching condi-
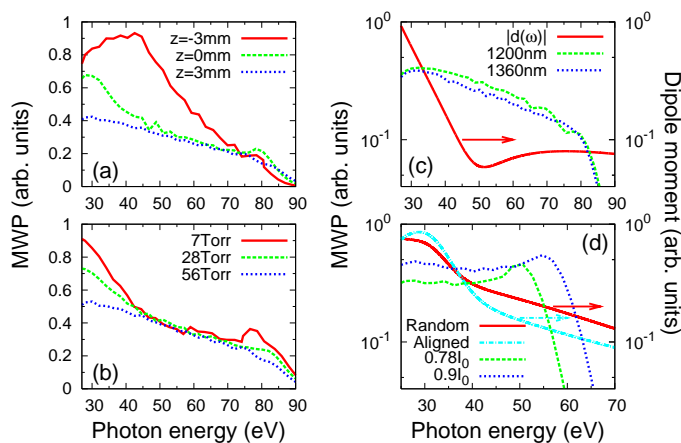
FIG. 3: (Color online) (a) Dependence of macroscopic wave packet $|W(\omega)|$ (MWP) on the position of the Ar gas jet with respect to laser focus; (b) Effect of gas pressure on MWP. The curves are renormalized such that they should fall on the same curve if perfect phase matching condition is fulfilled. (c) The MWP for the two lasers, and the magnitude of the PI transition dipole moment of Ar. (d) Same as in (c) but for N_2. The MWP is for two different laser intensities, and the averaged PI transition dipoles are for isotropic vs aligned molecules. The laser intensities are indicated where $I_0 = 10^{14}$ W/cm^2. See Text.

tion cannot be achieved for all the harmonics in a given experiment.

According to Eq. (1), the minimum in the HHG spectra can occur for different reasons. In Fig. 3(c), the MWP derived from Ar target using 1200 nm and 1360 nm lasers are shown. The two MWP's are quite similar but near 50 eV, they have slight different slopes. On the other hand, the PI transition dipole reveals a clear but broad Cooper minimum near 50 eV. Thus the broad minimum in the HHG spectra shown in Fig. 1(c) is due to the minimum in the PI transition dipole. To pin down the position of the "real" minimum, on the other hand, is not as easy since the minimum position can be modified somewhat by the MWP.

Similar analysis can be carried out on the HHG spectra of N_2 shown in Fig. 2. The averaged PI transition dipole indeed shows a rapid drop near 40 eV, which is due to the presence of a shape resonance [29] of N_2 in the lower energy. The rapid drop is more pronounced for aligned molecules than for random ones, see Fig. 3(d). For the MWP, under the same laser intensity, we have checked that they are the same for randomly distributed and aligned molecules. Thus it explains why the HHG from single-molecule response can be used to interpret how the intensity of each harmonic changes with pump-probe time delay in Le et al. [30]. However, the MWP changes more rapidly with laser intensity, especially for the longer wavelength laser used here. We note that the two MWP's in Fig. 3(d) have somewhat different slopes

near 40 eV. The multiplication of the MWP and the PI transition dipole results in a weak minimum in the observed HHG. The minimum would be more clearly seen if the molecules were better aligned. From Le et al. [13], the minimum in PI transition dipole changes rapidly with the alignment angle and the effect is severely averaged out when molecules are not well aligned. We further mention that the MWP in Figs. 3(c) and (d) are rather different. They are due to the large difference in the laser peak intensities used [25].

In summary, we demonstrated that experimental HHG spectra can now be accurately reproduced from ab initio calculations. The theory starts with the calculation of laser-induced dipoles from single atoms or molecules using the recently developed quantitative rescattering (QRS) theory [13, 14]. The propagation effect of the fundamental field and the harmonics in the medium is incorporated by solving the Maxwell's equations. We further showed that the simulated (and experimental) HHG spectra can be expressed as the product of a macroscopic wave packet and the photo-recombination transition dipole moment. The latter is a property of the target, and is independent of the lasers, nor of the propagation effect. This factorization makes it possible to extract target structure information from the experimental HHG spectra. It provides the needed theoretical basis for using HHG as ultrafast probes of excited molecules, such as those demonstrated recently [3]. Clearly, this work also opens up opportunities for the quantitative studies of the phases of HHG which are fundamental to the generation of attosecond pulses.

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