Mechanism of quasi-phase-matching in a dual-gas multijet array

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Quasi-phase-matching in a dual gas (Ar-H2) multijet array has recently been demonstrated to be a promising way to enhance the yield of high-order harmonics (HH). Here, we investigate the HH produced individually from these two gases under identical conditions. Our results indicate that the quasi-phase-matching results from the much lower recombination cross section of H2 as compared to that of Ar in the energy range of interest, rather than from full ionization of H2 by the driving laser as proposed previously.

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In the experiment, high-order harmonics were generated in a 1 mm long cell containing either Ar or H2 gas. The driving laser pulse was 30 fs long with spectrum centered at 780 nm. The generated XUV beam was focused to a Ne gas target, and the photoelectron spectrum was measured by a magnetic bottle electron energy spectrometer (MBES) [14]. Due to the nearly constant photoionization cross section of Ne in the extreme ultraviolet and soft x-ray wavelength regions, the photoelectron spectrum serves as an energy-shifted replica of the HH spectrum. HH with photon energies between 20 eV and 75 eV were measured for different gas pressures and driving laser intensities.

We first investigated the HH yield from Ar and H2 as a function of the target gas pressure, as shown in Fig. 1. The pressure inside the cell was the same for both Ar and H2 under the same backing pressure, as verified by comparing the absorption of the generated HH in an identical cell with the known absorption cross sections of Ar and H2 [15] and by gas flow simulations performed with FLUENT [16]. The laser intensity was 2.4(±0.5) × 1014 W/cm2, as determined from the cutoff harmonic generated in Ne gas. Because the MBES has a detection limit (∼10 000 electron counts/s), the pressure of the detection Ne gas jet was reduced for generation gas pressures above 20 torr, as indicated in the figure. Both Ar and H2 exhibit a quadratic increase of the HH yield with increased gas pressure, which indicates that both Ar and H2 are phase-matched as HH sources [17] when the pressure is less than 100 torr. The drop of the HH signal above 100 torr is likely due to the effects of the laser-produced plasma inside the generation target.

The results strongly indicate that the H2 target is not fully ionized by the laser pulse at this intensity; otherwise the HH signal from H2 would begin to decrease at much lower pressure than that from Ar. The similar roll-off pressure suggests that plasma density in the H2 target is nearly the same as that in the Ar target. However, the QPM is still achievable due to the pressure dependance of the H2 refractive index, which is different for the NIR driving laser and the HH. For partially ionized H2, the refractive index can be estimated as [17]

\[
n(\omega_q) \approx 1 + \frac{e^2 N}{2\varepsilon_0 m_e} \left( \frac{1 - P}{\omega_q^2 - q^2\omega_0^2} - \frac{P}{q^2\omega_0^2} \right),
\]

where

\[
P = \frac{2\varepsilon_0 m_e c^2}{\hbar^2} \omega_0^2, \quad \omega_q = \frac{q\omega_0}{\sqrt{1 + q^2}}.
\]
Fig. 1. (Color online) Dependence of the 27th HH on the generation gas pressure. (a) Ar and (b) H\textsubscript{2} can both be phase matched for pressure-length products up to approximately 100 torr × mm. The error is primarily due to determination of the pressure at low pressures.

where \( N \) is the H\textsubscript{2} molecular density which is related to the H\textsubscript{2} gas pressure, \( \omega_0 \) is the frequency of the NIR driving laser, \( P \) is the ionization probability, and \( \omega_r \) is the resonance frequency of the H\textsubscript{2} molecule. We can see that the refractive index for the NIR driving laser (\( q = 1 \)) is a function of gas pressure and ionization probability, while for HH (\( q \gg 1 \)), the refractive index \( n(q, \omega_r) \approx 1 \). The difference in the refractive index will lead to a phase difference between the NIR driving laser and HH after passing through the H\textsubscript{2} “buffer” jet. Furthermore, regardless of the ionization probability of H\textsubscript{2}, the phase difference is always controllable by adjusting the gas pressure, and the QPM can be realized with a phase difference of ±\( \pi \).

We then compared the HH generated from Ar and H\textsubscript{2} under different driving laser intensities. The pressure in the generation gas cell was 5 torr for both Ar and H\textsubscript{2}. The intensity within the focus was controlled using an iris, and was determined from the cutoff harmonic generated in Ne gas. Figure 2(a) shows the yield of the 19th harmonic (~30 eV) for different laser intensities from both experiment (symbols) and numerical modeling with the time-dependent density functional theory (TDDFT) (solid lines).

Traditionally many theoretical studies of multiphoton ionization (MPI) and HH generation (HHG) processes in molecules are based on various implementations of the strong-field approximation (SFA) and single-active-electron (SAE) model. However, while SFA-SAE-based models result in rather simple theoretical expressions, they fail to give quantitative agreement with more accurate theories. The discrepancy can be as large as several orders of magnitude. Besides other limitations, SFA-SAE-based theories usually deal only with the highest-occupied molecular orbital (HOMO) and neglect the multielectron dynamics of the target molecules. However, multielectron effects due to the electron exchange and correlation may be significant even when the inner electrons are strongly bound and are not excited by the driving laser field. Our theoretical method is based on the extension of TDDFT with proper long-range potentials (for a review, see [18]). The method takes into account the dynamic response of all the electron shells to the external fields and has been applied successfully to the nonperturbative study of MPI and HHG of atoms [19,20] and diatomic molecules [20,21] in intense laser fields. It is capable of reproducing the Cooper minimum in the HH spectrum of Ar as well as the contributions of multiple orbitals to the HH generated from molecular gases. In the present TDDFT study, we make use of the LB94 exchange-correlation potential [22] which describes accurately the electronic structure of both Ar and H\textsubscript{2}. Only hydrogen molecules with molecular axes aligned with the laser polarization were considered in the calculations.

In both the experiment and the simulation, the yield of the 19th HH is approximately an order of magnitude smaller for H\textsubscript{2} than for Ar, which is consistent with previous measurements [23]. The yield of the HH from Ar and H\textsubscript{2} are both observed to increase up to an intensity of ~3 × 10\textsuperscript{14} W/cm\textsuperscript{2}, where both gases have high ionization probability [13]. Above this intensity, ground-state depletion, plasma defocusing of the NIR laser in the generation target, and poor phase-matching

![Graph](a) Detection jet: 70 torr. Detection jet: 70 torr. -- fit exponent: 1.8184. ![Graph](b) Detection jet: 50 torr. Detection jet: 500 torr. -- fit exponent: 2.1896.

Fig. 2. (Color online) Scaling of HH with driving laser intensity. (a) Yields of 19th HH generated from Ar and H\textsubscript{2} increase with intensities up to ~8 × 10\textsuperscript{14} W/cm\textsuperscript{2} (symbols, experimental results; solid lines, TDDFT calculation results). The error is primarily due to determination of the laser intensity from the cutoff harmonic generated from Ne gas. (b) Ionization probabilities of Ar and H\textsubscript{2} from TDDFT calculation. The ionization of H\textsubscript{2} is reduced compared to Ar up to intensities of ~3 × 10\textsuperscript{14} W/cm\textsuperscript{2}. The laser pulse assumed in the simulation was a cosine-squared pulse with a 30 fs FWHM duration centered at 780 nm.
cause the HH yield to decrease. To verify this, the ionization probabilities for H2 and Ar were also simulated in the TDDFT calculations, as shown in Fig. 2(b). For laser intensities above $\sim 3 \times 10^{14}$ W/cm$^2$, the ionization probabilities of both H2 and Ar become close to unity. For lower intensities, the ionization probability of H2 is always smaller than that of Ar in spite of its lower ionization potential, which is consistent with previous experiments [12,13]. Therefore, H2 cannot be fully ionized at intensities for which HH generated from Ar is fully phase-matched in the multijet configuration.

In fact, the quasi-phase-matched buildup of HH observed by Willner [10] does not require that the H2 target be fully ionized, but only that the H2 gas does not efficiently generate or absorb the HH. In Fig. 2, we observed that the yield of the 19th harmonic generated in H2 was approximately an order of magnitude smaller than that of Ar for all intensities. Figure 3 shows the HH spectrum measured as a function of the driving laser intensity. As the laser intensity increases, the cutoff energy of harmonics generated from Ar increases up to the limit of the aluminum filter transmission ($\sim 73$ eV), which is well predicted by the semiclassical cutoff law for HHG [24]. At high intensities, HH signal from Ar could also be observed above the aluminum transmission edge by using a titanium filter. However, the yield of HH generated from H2 gas was observed to be very low above $\sim 60$ eV, and no harmonics could be observed extending above the aluminum transmission edge. In the entirety of the measured photon energy and laser intensity range, the strength of HH generated from Ar is always much higher than that from H2, which is required by dual-gas quasi-phase-matching.

The yield of HH is also closely related to the recombination cross section of the generation gas [25]. In Fig. 4, the measured HH spectra from Ar and H2 with laser intensity of $4 \times 10^{14}$ W/cm$^2$ are compared with their respective photoionization cross sections (PICS) [15], which are proportional to the recombination cross sections. We observe that the spectrum of the HH follows the PICS of the target atom. Small deviations of the HH spectra in Fig. 4(b) from the photoionization cross section shown in Fig. 4(a) are likely due to the nonuniform transmission of the aluminum filter and variations in the PICS of the Ar HH (upper) extends to the aluminum filter transmission edge. The HH signal from H2 is lower than that from Ar in the entire spectrum and laser intensity range.

FIG. 3. (Color online) HH spectrum as a function of the laser intensity. Ar HH (upper) extends to the aluminum filter transmission edge. The HH signal from H2 is lower than that from Ar in the entire spectrum and laser intensity range.

FIG. 4. (Color online) Cross-section dependence of HH yield. Comparison of (a) the photoionization cross sections with (b) the measured HH spectra. (c) Comparison of the cross-section ratio to the ratio of the spectrally resolved HH yields, eliminating the effects of the Al filter and detection gas. The error is primarily caused by the low signal levels of H2 HH above 60 eV.

Ne detection gas. However, these effects are removed by taking the ratio of the harmonic spectra from the two gases, which is in good agreement with the PICS ratio shown in Fig. 4(c).

This dependence of the HH yield on the PICS explains two features observed in the HH spectrum generated in H2. First, the reduced efficiency of H2 HH (as well as the low absorption necessary for quasi-phase-matching) is largely due to the small recombination cross section. The PICS of H2 is approximately an order of magnitude smaller than that of Ar in this energy range (except near the Cooper minimum at $\sim 46$ eV). Therefore, the HH yield of H2 is negligible compared to that of Ar. Second, the lack of a HH signal from H2 above $\sim 70$ eV is due to the reduced probability of recombination with such high photon energies. Whereas the PICS of Ar is nearly constant (or slightly increasing) above 50 eV, allowing observation of high-energy cutoff harmonics, the PICS of H2 decreases by nearly an order of magnitude between 50 and 75 eV. Because of these features, QPM could be achieved in Ar using H2 buffer gas, since the HH generated from H2 is far too weak to destructively interfere significantly with that generated in Ar.

In conclusion, we find that under the same driving laser intensity and gas pressure, the yield of HH generated from H2 gas is approximately an order of magnitude less than that from Ar. We propose that the relatively small recombination cross section of H2 is responsible for the decreased yield of HH. Overall, H2 exhibits low absorption of XUV light, low HH conversion efficiency, and tunable refractive index for the NIR driving laser, which allow it to be used as a passive medium for QPM of high-order harmonics generation. Although differing ionization rates may contribute to making this QPM scheme effective for certain gas pairings, such as He-H2, it is not a general requirement. The proposed mechanism for QPM extends the usefulness of the dual-gas multijet array to gas
pairs other than Ar-H₂, for which the PICS of the buffer gas is low in the spectral range of interest.

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