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Ellipticity dependence of atomic and molecular high harmonic generation

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Abstract. High harmonic generation is compared in the dependence on the ellipticity of the fundamental laser radiation for an atomic and a molecular system. In particular argon and nitrogen are compared employing molecular beams and intense (3×10^{14} W/cm^2) and ultrashort (80 fs) 800 nm laser pulses. It turns out that for all the harmonics under investigation (H5, H13 and H21) the harmonic yield decreases slower with the ellipticity for the molecule than for the atom. This indicates differences in atomic and molecular high harmonic generation.

PACS. 42.65.Ky Harmonic generation, frequency conversion – 32.80.Wr Other multiphoton processes

1 Introduction

High harmonic generation has attracted a substantial amount of attention during the last decade. With the generation of radiation in the water window (2.3–4.4 nm) [1–3] its potential for the realization of coherent sources in the vacuum- and extreme-ultraviolet spectral regions (VUV, XUV) has been demonstrated. Of particular interest is the time structure of harmonic pulses: only recently the pulse duration of a high harmonic was determined in cross-correlation measurements to lie in the attosecond time-regime [4,5] and trains of attosecond-pulses have been observed from high harmonic generation [6].

The process of high harmonic generation is understood in the framework of the semi-classical three step model [7–9]. In the first step an atom or molecule is ionized by the strong electric field of the laser through tunnel-ionization. The free electron is then accelerated away from the ionic core in the oscillating laser field, however, when the laser-electric field changes sign, the free electron is moved back to the core and can in a third step eventually recombine. The energy which the electron picks up by the acceleration in the laser field (plus the ionization potential of the atom) is then emitted in a harmonic photon. This model was successfully applied to the description of high harmonic generation and was also formulated quantum-mechanically [9]. It can predict the highest photon energy by the so-called cut-off-law as well as the shape of harmonic spectra.

One assumption commonly made in the three step model is that the potential of the core does not influence the trajectory of the free electron. The influences of the system are the transition from the ground state to the continuum (tunnel ionization) and to the recombination process. The harmonic generation process, however, is dominated by the second step, where the electron is accelerated in the continuum [8,9]. Therefore we should not expect to see huge differences between atomic and molecular high harmonic generation.

There have been several experiments performed until now, comparing high harmonic generation in different atomic and molecular gases [10–16]. In [10] the intensity dependence of harmonic generation from several molecules was compared with that from rare gases with respect to the three parameters field-free ionization potential, mass and static average polarizability. It turned out, that the static average polarizability is an important factor, determining the dependence of harmonic generation to the laser intensity. A similar study was done by Lyngå *et al.* [13] comparing a huge range of different diatomic as well as polyatomic molecular gases with rare gases. It turned out that the molecules show very similar behaviour to rare gases.

In this paper we present a comparison between the ellipticity dependence of high harmonic generation in an atom and a molecule, argon and nitrogen. These two gases have been extensively studied in high harmonic generation and show very similar behaviour like similar cut-off and efficiency due to similar ionization potentials (15.8 eV and 15.6 eV, respectively) and static average polarizabilities (1.7×10^{-24} cm^3 and 1.6×10^{-24} cm^3,

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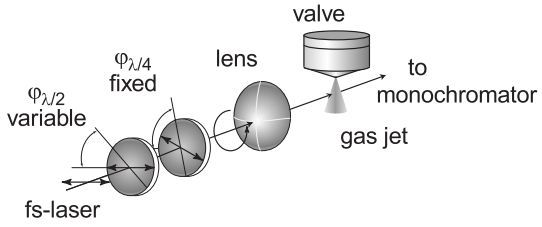

Fig. 1. Overview of the experimental setup.

respectively) [10,11,13]. Further similarities with respect to harmonic generation can be found in [17], where the third order susceptibility was determined by third order harmonic generation at $\lambda = 1\,055$ nm. The reported values were $\chi_0^{(3)}(\mathrm{Ar}) = 23.5 \times \chi_0^{(3)}(\mathrm{He})$ and $\chi_0^{(3)}(\mathrm{N_2}) = 21.1 \times \chi_0^{(3)}(\mathrm{He})$. Differences in the behavior between those two substances have until now only been observed in the polarization state of the generated harmonics [18]. In our work we find differences between the atomic and molecular response in the dependence of the total harmonic yield on the ellipticity of the driving field.

Measurements of the harmonic yield depending on the ellipticity of the fundamental laser radiation have been performed as a stringent test for the validity of the semiclassical three step model [19–21]. Since for harmonic generation the free electron wave-packet has to return to the core, a slight ellipticity in the fundamental laser beam will drastically reduce the efficiency of the process. Effects like phase-matching and propagation in the medium show a relatively weak dependence on the ellipticity of the fundamental laser radiation. Therefore measurements of the ellipticity dependence of high harmonic radiation are used to investigate the single-particle response isolated from these effects [22]. Measurements of ellipticity dependencies also served to illuminate the basic mechanisms in nonsequential double ionization [23,24] which turned out to be a closely related process to high harmonic generation.

2 Experimental setup

The experimental setup is shown in Figure 1. We used a commercial chirped pulse amplification (CPA) Ti:sapphire laser system from Spectra Physics. It produces laser pulses at a center wavelength of 800 nm, with a pulse duration of 80 fs and an energy of 900 μJ at a 1 kHz repetition rate. The laser is focused with a 300 mm focal length lens into a vacuum chamber. We determined the focused laser intensity to be 3×10^{14} W/cm^2 by measuring the ion yield intensity dependence of different noble gases with a time-of-flight mass-spectrometer. By comparing the results of these measurements with calculations of the ion yield based on the well known Ammosov-Delone-Krainov-formula for tunneling ionization (ADK) [25], we could determine the peak intensity in the focus.

The laser beam is originally linearly polarized. In order to change the ellipticity of the laser beam, we used a setup consisting of a half-wave-plate and a quarter-wave-plate as shown in Figure 1. Both wave-plates were 0^{th}-order wave-plates for 800 nm. The half-wave-plate was placed on a rotating mount to change the orientation of the linearly polarized electric field vector whereas the orientation of the quarter-wave-plate was kept fixed. The orientation of the half-wave-plate was controlled by a computer. By this we were able to change the ellipticity continuously between 0 and 1.

The focus of the laser beam was placed very close to the exit of a piezoelectric pulsed valve which was backed by either Ar or N_2 at a pressure of 2.0 bar, yielding an atomic/molecular beam. This valve was built according to the design given in [26] and is capable of operating at the repetition rate of 1 kHz which matches the repetition rate of our laser system. A more detailed description of the application of this valve to high harmonic generation is given in [27]. The atomic/molecular beam of the gases ensures an investigation of single events in contrast to gas cells.

To detect the radiation produced in the target beam we used two different monochromators: a home-built Seya-Namioka monochromator, which allowed us to detect the low order harmonics (H3–H17), and a grazing incidence monochromator (Jobin Yvon, LHT30) for the higher order harmonics (above H11). The spectrally resolved VUV-radiation was converted to visible fluorescence with Na-salicylate as a scintillator and detected with a photomultiplier tube. Data were collected with a Boxcar integrator and stored to a computer.

3 Results

The monochromator was set to the wavelength of a harmonic and for every setting of the half-wave-plate the signal was averaged over at least 2 500 laser shots. The half-wave-plate was moved either in steps of 1° or 0.5° for a full rotation. That means that the ellipticity was changed 16 times between 0 and 1 in a single scan. An example of such a measurement is shown in Figure 2 where the solid line shows the harmonic signal at H13 from Ar *versus* the angle of the half-wave-plate. As the half-wave-plate is turned, the polarization of the fundamental is changed between linear and circular and the signal follows these changes. The ellipticity for every angle of the half-wave-plate is given by the dashed line.

In addition to the change in the signal with the ellipticity, there appears a periodic change in the height of the maxima corresponding to linear polarization. This is due to the grating efficiency which is different for *s*- and *p*-polarized light. From the different signal levels corresponding to *s*- and *p*-polarized light we can deduce this difference in the grating efficiency and use this in the data analysis to correct our data for the influence of the grating.

To analyze the data we averaged over all points which belong to the same ellipticity, taking the grating efficiency into account as described above. From that we obtained the harmonic intensity *versus* the ellipticity, which was then normalized to one in order to compare different scans.

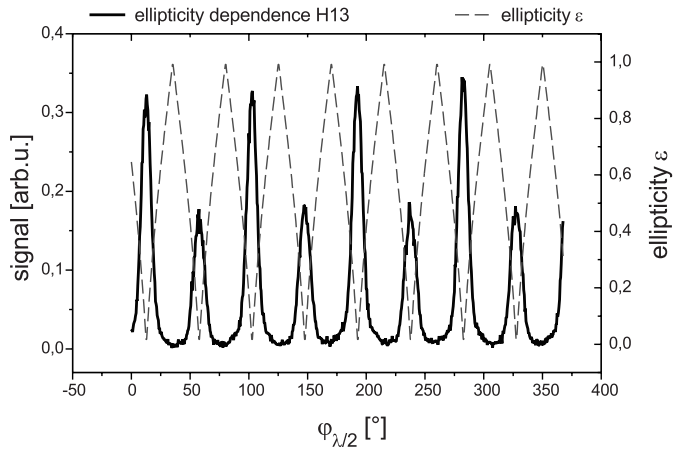

Fig. 2. Measurements of the H13-intensity from Ar plotted *versus* the angle $\varphi_{\lambda/2}$ of the half-wave-plate with respect to the original orientation of the linear laser polarization. The solid line is the measurement of the ellipticity dependence. The dashed line shows the value of the ellipticity for every angle of the half-wave-plate.

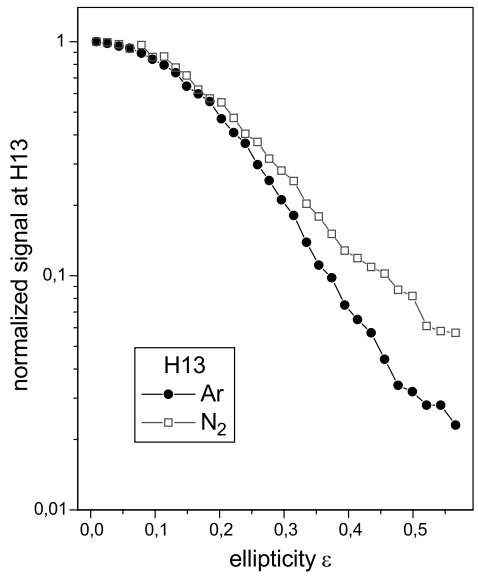

Fig. 3. Ellipticity dependence of H13 in Ar (black circles) and N_2 (grey rectangles).

The result of this procedure is shown in Figure 3. In this figure two measurements of the ellipticity dependence are shown, where between the different measurements only the gas was changed from Ar to N_2, all other experimental parameters were kept constant. Data are shown for ellipticities below 0.6. At these values of the ellipticity the harmonic signal dropped beyond the noise level of our experimental setup. We determined the dominant contribution to this noise to be stray light in the monochromator. In order to clarify the contribution of this stray light to the measurements of the ellipticity dependencies we repeated the experiments with the monochromator set to a wavelength away from the harmonics and subtracted this background from the data. Since this background subtraction

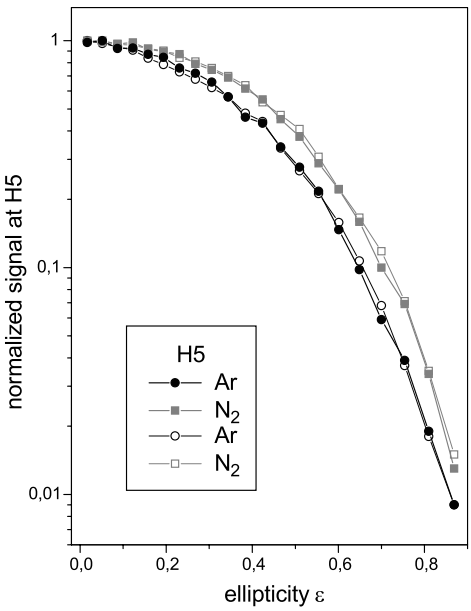

Fig. 4. Ellipticity dependence of H5 in Ar (black circles) and N_2 (grey rectangles). Shown are consecutive measurements, where only the gas was changed in between.

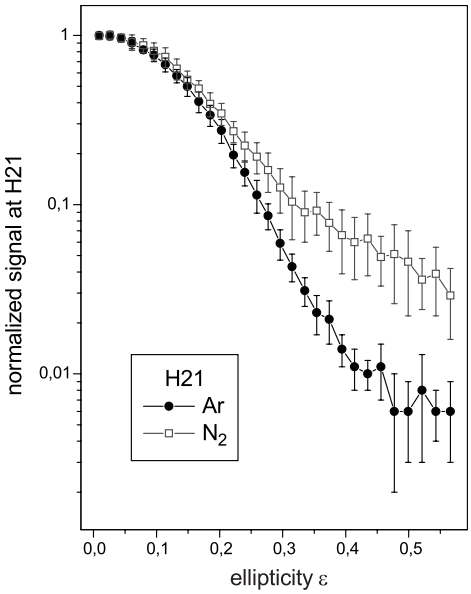

Fig. 5. Ellipticity dependence of H21 in Ar (black filled circles) and N_2 (grey open rectangles).

did not change the general features of our measurements – the molecule showing a weaker dependence on the ellipticity – we can rule out that the observed effect is due to stray light and we decided to show the raw data without any background subtraction.

In Figure 3 the curves for N_2 lie clearly above the Ar-data. To check whether this finding is a general feature we repeated this experiment for a low order harmonic – H5 shown in Figure 4 – and for a harmonic lying further in the plateau – H21 shown in Figure 5. To ensure the reproducibility of our measurement, four measurements are

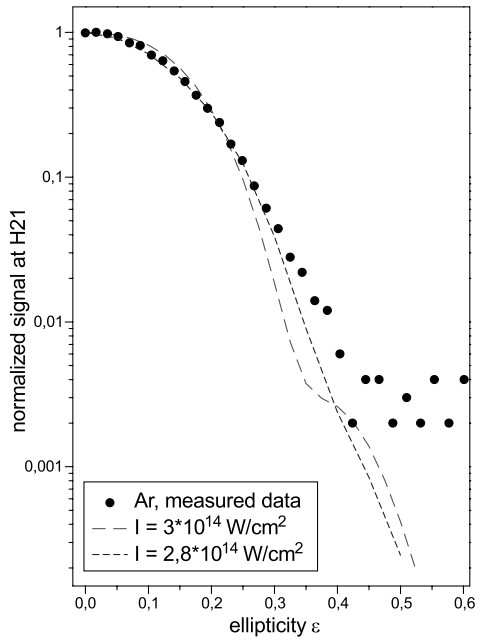

Fig. 6. Measurement (black filled circles) and simulations (dashed lines) of the ellipticity dependence of H21 in Ar. The simulations have been done for two different laser intensities.

shown in Figure 4. These correspond to successive runs, where only the gas was changed in between. Note that with our peak intensity of 3×10^{14} W/cm^2, we already partially entered the barrier-suppressed-ionization [28] regime, where perturbation theory for low-order harmonics is not valid. Therefore, the ellipticity dependence of H5 in argon is slightly weaker than perturbation theory predicts. In Figure 5 error bars are included for the ellipticity dependence of H21. These error bars are determined by averaging over all points of the same ellipticity.

In Figure 6 the ellipticity dependence of H21 in Ar is shown again together with simulations of the single-atom response. For this comparison we subtracted the stray-light background as described earlier. The simulations have been done in a fully quantum-mechanical description [29], valid in the tunneling limit and based on the quantum-mechanical model for high harmonic generation given by Lewenstein and coworkers [9]. In our simulations we used peak laser intensities of 3×10^{14} W/cm^2 (black long-dashed line) and 2.8×10^{14} W/cm^2 (grey short-dashed line). For the numerical calculations we make the same approximations as in [29] such as restricting the integration over time to only four laser cycles and considering only the lowest eight dominant components in the Fourier-transform of the time-dependent dipole moment.

The simulated curves exhibit a strong dependence on the laser intensity. This is related to oscillations of the dipole strength due to quantum interference effects between different electron trajectories. Also the knee structure in the simulation for $I = 2.8 \times 10^{14}$ W/cm^2 at around $\varepsilon = 0.35$ is due to quantum interference. The influence of quantum interference effects has been discussed by Antoine *et al.* in [29].

For the simulations in Figure 6 propagation effects have not been included in the calculations. To compare the experimental data with the theoretical curves one has also to keep in mind that the calculations have only been done for two different laser intensities. However, since the measured signal contains contributions from different parts of the spatial laser beam profile we should expect an averaging over several intensities. To take this averaging into account we would need to consider the propagation of the fundamental and harmonic fields in the gas, which could be done according to [29] but has not been included here.

However, by comparing the measured with the calculated data for Ar, we can get a hint on the validity range of our measurements. At ellipticities larger than $\varepsilon = 0.5$ the stray light background becomes dominant and we should restrict our discussion to ellipticities $\varepsilon \leq 0.4$.

For all three harmonics shown here one can see that N_2 does always exhibit a slower decrease with the ellipticity of the fundamental radiation than Ar. We also tried to repeat this comparison for a harmonic lying in the cut-off, namely H27. However, since the signal level for this harmonic was already relatively low, we were not able to obtain the dynamic range needed to measure the ellipticity dependence for ellipticities above 0.2.

This comparison was also repeated for different stagnation pressures of argon and nitrogen. For all different pressures we found the molecule to exhibit a weaker dependence on the ellipticity of the fundamental. Here we show only data taken with a stagnation pressure of 2.0 bar.

Additionally we studied the influence of phase-matching on the ellipticity dependence by repeating the measurements for various relative positions of gas-jet and laser focus, which is a crucial parameter for phase-matching of the high harmonic generation process [30]. These measurements showed that the ellipticity dependence does not vary with the relative position of gas-jet and laser focus at all. Therefore measurements of the ellipticity dependence are related to the single-particle response and not influenced by phase-matching effects as has already been stated in [22].

4 Discussion

The results of our measurement indicate that the probability for harmonic generation at a given ellipticity is always higher for the N_2-molecule than for the Ar-atom. Similar results are reported in [14] where the ellipticity dependence of xenon atoms, benzene and *n*-hexane molecules was compared using laser pulses of 70 fs duration. In these experiments the harmonic signal from the molecules was higher than from the atoms, like we observed in our measurements. This is in contrast to an earlier experiment with somewhat larger laser pulse duration (240 fs) [16], where the atom (xenon in this case) exhibited a slower decrease of the harmonic signal with ellipticity than the investigated benzene molecules. This finding was explained in [16] by fragmentation of the molecules and harmonic generation from fragments rather than molecules. The fact

that we observe the molecular curves of the ellipticity dependencies always above the atomic ones and the pulse duration of 80 fs used in the experiments described here being similar to the 70 fs laser pulses used in [14] indicates that in our experiment the harmonics are generated by the molecules and fragmentation can be ruled out. In [14] the observed phenomenon is explained by the effective cross-section for recombination being larger in the molecule than in the atom. This implies in the molecular case additional pathways for the electron to recombine with atoms different from its parent atom.

Another possible explanation can be found in [31], where selection rules for high harmonic generation are derived based on the symmetry of the species used to generate high harmonics. The well-known fact, that only odd harmonics can be produced from atoms within a linearly polarized laser field is reproduced there. Also the case of a molecule exhibiting C_N-symmetry in a circularly polarized laser field is studied. It turns out that in this case the single-particle response allows for the generation of specific harmonics only. Since the single-particle response allows for the generation of harmonics with circularly polarized light in the case of a C_N-symmetric molecule but not in the case of an atom, we expect a slower decrease of the harmonic signal with the ellipticity of the fundamental laser light for the molecules than for the atoms.

In that sense measurements of the ellipticity dependence are not only sensitive to the single particle response but also to the symmetry properties of the particle. However, in our experiment the molecules are randomly oriented and therefore we should expect an averaging over all orientations. Further experiments in this direction using aligned molecules will be needed to investigate the influence of symmetries on the harmonic generation process.

5 Conclusion

In conclusion we have studied the ellipticity dependence of atomic and molecular high harmonic generation by measuring the high harmonic yield of argon and nitrogen. For all harmonics under investigation (H5, H13 and H21) we found that the molecular response shows a weaker dependence on the ellipticity of the driving field than the atomic one. This reveals differences between atomic and the molecular high harmonic generation.

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