

Influence of ellipticity on harmonic generation

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We present results of experiments testing the influence of elliptical polarization on the production of high-order harmonics. Experiments were conducted both with a 600-nm, 1-psec dye laser and with an 825-nm, 140-fsec Cr:LiSrAlF₆ (Cr:LiSAF) laser system, over a wide range of intensities and target gases (xenon, argon, and neon), using a detection system with a dynamical range of more than three orders of magnitude. The decrease of the harmonic strength with the ellipticity of the pump beam is rather slow for the low-order harmonics, and becomes much steeper for the high-order harmonics. We compare some of these data with the predictions of lowest-order perturbation theory.

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High-order harmonic generation exhibits a characteristic spectrum consisting of a decreasing efficiency for the first few harmonics followed by an extended plateau, or region of essentially constant conversion efficiency, ending with a sharp cutoff. It has been shown that the energy at which the cutoff occurs in the single-atom response is well approximated by the expression $I_P + 3U_P$, where I_P is the atomic ionization potential and $U_P(\text{eV}) = 9.33 \times 10^{-14}[I(\text{W}/\text{cm}^2)][\lambda^2(\mu\text{m}^2)]$ is the ponderomotive energy of the electron in the laser field [1]. Classical approaches [2,3] as well as a simple quantum-mechanical theory [4] have been used to give some physical insight into this formula. In this picture, the electron tunnels through the atomic potential, lowered by the presence of an intense laser field. Its motion in the field is essentially that of a free electron and can then be treated classically. If the electron trajectory is one that returns to the nucleus, the electron may recombine to the ground state and emit a harmonic photon. If the trajectory is such that the electron does not return to the nucleus, the atom will simply ionize and no harmonics will be produced. One way of controlling the trajectory of the electron is to alter the polarization of the incident laser field, making it elliptical rather than linear [5,6].

In this Rapid Communication, we investigate the influence of the ellipticity of the incident laser beam on harmonic generation. The experiments were performed in different regimes defined by the tunneling parameter, $\gamma = (I_P/2U_P)^{1/2}$. For $\gamma > 1$, multiphoton processes dominate while $\gamma < 1$ denotes the regime where tunneling is the primary ionization mechanism and where harmonic generation is well described by the quasiclassical picture described above. By varying the laser frequency,

the atomic system, and the laser intensity, we can probe both of these regimes.

We use two different laser systems. The first one is a short pulse dye laser consisting of a synchronously pumped linear cavity dye oscillator which is amplified in three stages. The amplifiers are pumped by 75-psec pulses at 532 nm from a Nd:YAG (neodymium-doped yttrium aluminum garnet) regenerative amplifier that allows for a contrast of greater than 10^5 between the amplified pulse and the amplified spontaneous emission (ASE) background. Up to 4 mJ per pulse tunable from 570 to 620 nm with a 1-ps pulse duration are produced [7]. Here the laser is operated at 600 nm (2 eV) at a 5-Hz repetition rate and focused by a 30-cm lens to a 1.5-times diffraction-limited spot to produce intensities up to 10^{14} W/cm². Due to the relatively low intensity only xenon is used as a target gas. The second laser is a Cr:LiSAF chirped pulse amplification system. A pulse from a mode-locked Ti:sapphire oscillator is stretched temporally and used to seed a Cr:LiSAF regenerative amplifier. The output is then amplified in a 4-mm and two 9-mm Cr:LiSAF amplifiers and recompressed producing up to 150 mJ in a 140-fsec pulse at 825 nm (1.5 eV) [8]. With a 1-m focusing lens and a 3-times diffraction-limited spot, intensities greater than 10^{15} W/cm² are achievable and neon, argon, and xenon are used as the harmonic generating medium. The repetition rate is 0.3 to 1 Hz depending on the energy used.

The dye laser is vertically polarized by using a polarizer after the amplifiers. The Cr:LiSAF laser is horizontally polarized, and this has been verified by using a thin-film polarizer. Both lasers are passed through a broadband (zero-order) quarter-wave plate (centered at 600 nm or

at 825 nm) allowing us to vary the polarization continuously from linear to circular. They are focused below the nozzle of a pulsed gas jet, which produces an atomic pressure of 20 Torr. The harmonics produced are spectrally dispersed by a flat-field monochromator (9–90 nm) consisting of a toroidal mirror and a variable groove density plane grating, both coated in gold. The signal is detected with an electron multiplier over a large dynamic range (more than three orders of magnitude) and data acquisition is PC controlled [9]. We have examined the influence of the polarization on the grating efficiency by recording the signal for a given harmonic for both vertically and horizontally polarized light. The change in signal level is less than a factor of 2 and within our experimental error bar for this measurement.

The experiment consists of recording the harmonic signal as a function of ellipticity, defined as the ratio of the y (minor axis) to the x (major axis) components of the laser electric field, for a fixed laser intensity. All data presented in Figs. 1–4 are normalized to the number of photons obtained for linear polarization and correspond to an average of 30 shots with a strict selection in energy (of $\pm 15\%$). The same horizontal and (logarithmic) vertical scales are used in order to make the comparison between the different results straightforward. We also indicate in the figure captions the normalization factors used for all the harmonics. The results presented here in xenon at 600 and 825 nm and in argon and neon at 825 nm are at intensities close to the saturation intensities for these gases and laser frequencies. We have performed additional measurements at different intensities, but the dependence of the signal with ellipticity was found to be very weakly dependent on the laser intensity over the range studied.

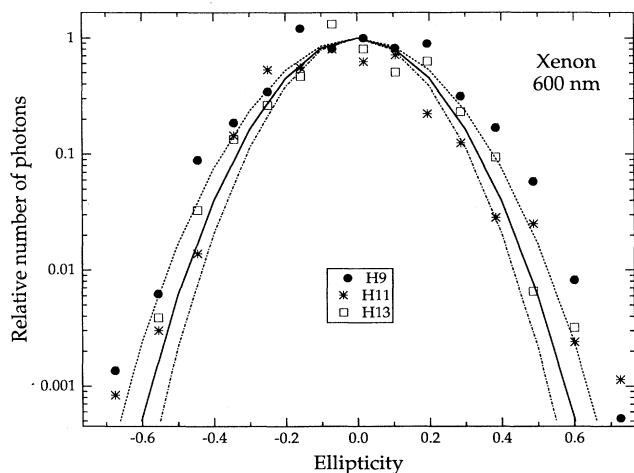


FIG. 1. Harmonics produced in xenon with a dye-laser system operating at 600 nm as a function of ellipticity, normalized to 4×10^8 photons at the 9th harmonic, 2×10^7 photons at the 11th harmonic, and 4×10^5 photons at the 13th harmonic. The intensity is 6×10^{13} W/cm². The lines indicate the decrease in harmonic production with increasing ellipticity as predicted by lowest-order perturbation theory for the 9th (dashed line), 11th (solid), and 13th (dot-dashed) harmonics.

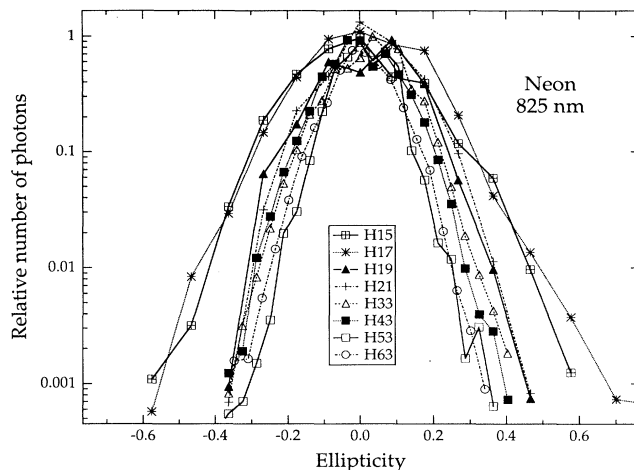


FIG. 2. Harmonics produced in neon with the Cr:LiSAF laser system as a function of ellipticity, normalized to 3×10^6 photons at the 15th, 19th, and 21st harmonics, 3.7×10^6 photons at the 17th harmonic, 6×10^6 photons at the 33rd harmonic, 4×10^6 photons at the 43rd harmonic, 2×10^6 at the 53rd harmonic, and 1.5×10^6 photons at the 63rd harmonic. The intensity is 10^{15} W/cm².

Figure 1 presents the results obtained with the 600-nm dye laser in xenon for the 9th, 11th, and 13th harmonics (the only detectable harmonics). The intensity is estimated to 6×10^{13} W/cm², so that $\gamma \sim 1.7$. In this multiphoton regime, a relatively weak dependence on ellipticity is observed. An ellipticity of 0.4 is required for a one order of magnitude decrease in harmonic strength and some signal is detected out to an ellipticity of nearly 0.7. The data are compared to the prediction of lowest-order perturbation theory [10,11],

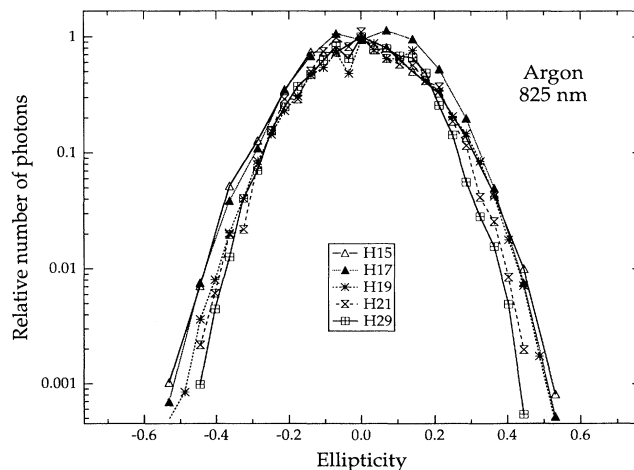


FIG. 3. Harmonics produced in argon with the Cr:LiSAF laser system as a function of ellipticity, normalized to 2×10^8 photons at the 15th harmonic, 3×10^8 photons at the 17th harmonic, 4×10^8 photons at the 19th harmonic, 5×10^8 photons at the 21st harmonic, and 1.3×10^8 photons at the 29th harmonic. The intensity is 3.5×10^{14} W/cm².

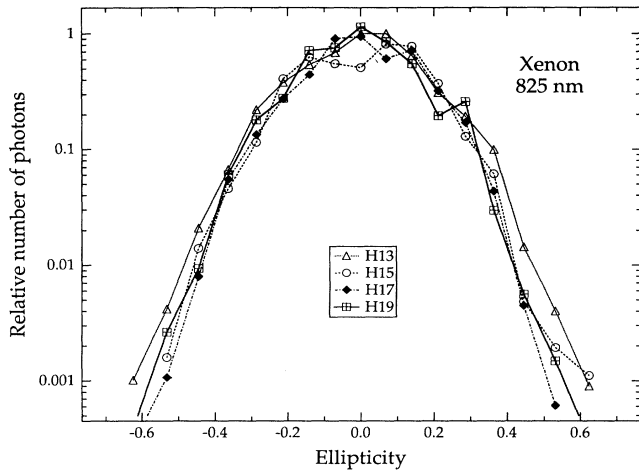


FIG. 4. Harmonics produced in xenon with the Cr:LiSAF laser system as a function of ellipticity, normalized to 3.5×10^9 photons at the 13th harmonic, 2.2×10^9 photons at the 15th harmonic, 5×10^8 photons at the 17th harmonic, and 3.5×10^7 photons at the 19th harmonic. The intensity is 1.2×10^{14} W/cm²

$$I_q \propto \left(\frac{1 - \epsilon^2}{1 + \epsilon^2} \right)^{(q-1)}, \quad (1)$$

where ϵ is the ellipticity as defined above and q is the harmonic order. The results are shown in Fig. 1 by the dashed (9th harmonic), solid (11th harmonic), and dot-dashed (11th harmonic) curves. The experimental data are consistent with the prediction of lowest-order perturbation theory, being in general slightly higher. In this regime, the polarization of the harmonics is expected to follow that of the pump beam [10].

Figures 2–4 show harmonics produced in neon, argon, and xenon with the LiSAF laser system. The experimental data have been connected by lines for the sake of clarity. The intensities are respectively 10^{15} W/cm², 3.5×10^{14} W/cm², and 1.2×10^{14} W/cm², i.e., $\gamma \sim 0.41$, 0.59, and 0.89 (the ionization energies are 21.6, 15.8, and 12.1 eV, in neon, argon, and xenon respectively). In this high-intensity regime, lowest-order perturbation theory becomes inadequate. In neon, for the harmonics well above the ionization threshold, the process of harmonic generation should be well described by the quasiclassical picture, where the electron first tunnels out, then moves freely in the laser field, and finally recombines to the ground state [2–4]. This theory predicts a strong dependence of the harmonic strength on ellipticity, a small fraction of the circularly polarized electric field being sufficient to prevent the electron from returning to the nucleus. In contrast, the low-order harmonics in xenon should be described by (nonperturbative) multiphoton processes, such that the charge density remains close to the nucleus, and bound states play an important

role [12].

The 15th, 17th, 19th, 21st, 33rd, 43rd, 53rd, and 63rd harmonics obtained in neon are presented in Fig. 2. The distributions become narrower as the order increases, with a rather pronounced transition between the 17th and the 19th harmonics. Above the 19th order, the signal level begins to drop off with less than 0.1 ellipticity, and has decreased by more than one order of magnitude by an ellipticity of 0.2. Approximately the same dependence on ellipticity is noted over higher harmonic orders, from the 19th to the 63rd order. The highest harmonic observed with linear polarization is the 85th, but without enough signal-to-noise ratio to study the influence of ellipticity in a meaningful manner. Lower orders, 13th–17th, exhibit a weaker dependence, requiring at least 1.5 times more ellipticity to see the same decrease in signal as exhibited by the higher orders. Note that lowest-order perturbation theory in general predicts a steeper dependence than that exhibited here, in particular for the high harmonics.

The 15th, 17th, 19th, 21st, and 29th harmonics obtained in argon are displayed in Fig. 3. The distributions become slightly narrower with increasing order, but there is no abrupt transition between two following harmonics as observed in neon. Finally, the results obtained for the 13rd, 15th, 17th, and 19th harmonics in xenon are shown in Fig. 4. Only a weak dependence on ellipticity is observed. These data also show a dependence with ellipticity that is weaker than the perturbative predictions, and evidently the discrepancy increases with the process order.

The results obtained in the three rare gases for low-order harmonics, 15th and 17th, though at different laser intensities, are extremely similar. Neon and, to a lesser extent, argon show a definite narrowing of the distribution of harmonic strength as a function of ellipticity from the 19th harmonic, which is not observed in xenon. These data illustrate a transition region where the degree of ellipticity of the incident laser begins to have a large effect on the production of the high harmonics. Note that for those high harmonics, i.e., beyond the 19th, the dependence with ellipticity is practically independent of the harmonic order. A more detailed comparison with theoretical calculations performed in this nonperturbative regime and including the effect of propagation will be presented in a forthcoming paper.

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