Polarization of the 61st harmonic from 1053-nm laser radiation in neon

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We report the polarization measurement of a very high order (59th and 61st order, ~17 nm) harmonic. A Mo-Si multilayer mirror is used as a polarizer with reflectivity of 60% and polarization analyzing power close to unity around a wavelength of 17 nm. We observe that for these high harmonics in Ne there is no rotation of the polarization ellipse with respect to the fundamental laser polarization, even though the harmonics are in the plateau. For linear and slightly elliptical laser polarization the harmonics still remain linearly polarized. These findings are supported by a full theoretical simulation, which includes spatiotemporal integration and phase matching of the emitted harmonic radiation. [S1050-2947(98)07803-2]

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I. INTRODUCTION

During the last decade, owing to the development of intense ultrashort pulsed lasers, it has become possible to construct new x-ray ultraviolet (XUV) sources with unique properties. A prominent example is high-order harmonic radiation, which appears when intense laser light is focused into a gas. These harmonics are generated at odd multiples of the incident frequency (see, e.g., [1–3]). This phenomenon is characterized by a strong intensity decrease for the first few harmonic orders, followed by a broad range of harmonics only sightly decreasing in intensity, known as the ''plateau.'' At the blue end of the spectrum there is a characteristic sharp decrease (cutoff) depending on the laser intensity and the electronic binding energy in the medium [4].

Theoretical modeling of the process must include on one hand the atomic emission of the harmonics and on the other hand their subsequent propagation through the medium. The propagative part requires the solution of Maxwell's equations with source and diffractive effects and has been driven primarily by the group of L'Huillier and co-workers [5]. The atomic high-harmonic generation (HHG) is a much more difficult problem to treat in principle since the quantum mechanics of a driven multielectron system must be solved. Fortunately, however, HHG can be well described qualitatively and even quantitatively by considering single-active-electron models [6]. Even simpler ''two-step'' models have grasped the essential physics of the mechanism that for the HHG reduces to a quasifree (semiclassical) electron driven by the field and interacting punctually with the atomic core (see the references in [5]).

Presently the high-harmonic generation (HHG) becomes more and more interesting for applications [7,8]. Therefore a full characterization of the emitted HHG light is required. Evidently the absolute total photon flux in the harmonics is a key parameter [9], which it is imperative to optimize. On the way to even further control and possibly finer understanding of the HHG process, the harmonic yield and polarization in its dependence on the driving laser polarization is analyzed. This provides a much more detailed and quantitative test on the current theoretical models. First results have been obtained in [10,11] and a detailed theoretical [5] and experimental [12] study has been performed. The measurements in these studies have, however, been restricted to harmonic orders up to the 33rd and the polarizers used had a contrast ratio of about 0.9.

For our experimental study, we have used a polarizer consisting of a multilayer mirror, which has a near-optimal (unity) analyzing power. We selected essentially a single very high harmonic (the 61st) without the use of extra dispersive elements (grating) in the harmonic beam, which could change the harmonics' state of polarization. It has been observed [13,14] that the ellipticity dependence of the total harmonic yield follows the perturbative law for harmonic orders up to about 31, while the 61st harmonic in its ellipticity dependence falls off much slower than the perturbative prediction, no longer following this simple scaling.

We have also performed a complete theoretical numerical simulation of the experiment. To this extent we have developed an implicit, flux-conserving radiation propagation algorithm (assuming two-dimensional symmetry around the laser beam propagation axis) [15] and used various atomic models to describe the source of harmonics. We obtain very good agreement between theory and experiment using as the atomic model the three-dimensional (3D) delta-potential [16].

II. EXPERIMENTAL ARRANGEMENT

We used as pump source the front end of the Max-Born-Institute's high intensity CPA laser system [17]. It consists of a titanium:sapphire mode-locked oscillator (Tsunami - type, Spectra Physics), a double pass grating-telescope stretcher, a titanium:sapphire regenerative amplifier (Spectra Physics), two Nd:glass amplifiers and a grating compressor. It delivers pulses with 1053 nm, 20 mJ, and 700 fs. A 60-cm lens focused the pulses into a neon gas jet with a gas density of about 2×10^{18} cm^{-3}. The gas nozzle (series 9, General Valve) is mounted in a vacuum chamber. The focus diameter was about 65 μm.

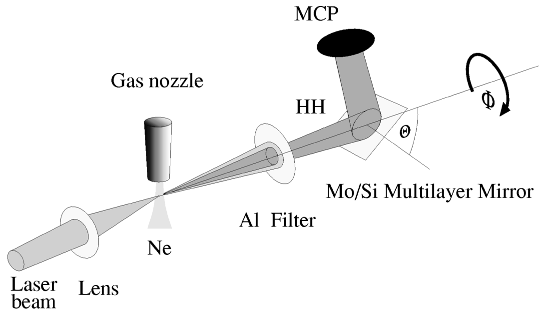

FIG. 1. Experimental setup for polarization measurement. Ne: neon gas jet at the laser focus. HH: high harmonics' beam. MCP: multichannel plate detector.

For our first measurements, reporting total photon yields we have used a more conventional spectrometer arrangement, consisting of a toroidal mirror, a plane flat-field grating and a special beam dump for the intensive IR laser radiation, placed in the zeroth order of the grating. A two-stage microchannel plate and phosphor screen arrangement converts the vacuum ultraviolet radiation into green light, which was detected by a charge-coupled device camera.

Both the investigation and possible applications of the HHG radiation require high reflectivity optics in the short wavelength range. Multilayer optics have already proven to be a powerful tool for focusing [18] as well as for polarization analysis [19]. Thus, for our polarization measurement, we used a multilayer reflector, in contrast to the previous experiments [11,12]. Use of the multilayer mirror has three advantages: (1) There is no need for a dispersive element (grating), which can alter the polarization of the harmonic radiation and whose characterization would be a source of error. (2) The reflectivity of our mirror, R_p, for p-polarized light is close to zero at the wavelength of the 61st harmonic considered and as a result the multilayer has an analyzing power, which is practically equal to unity (see below). (3) The high reflectivity of 60% for a spectral range from 13 to 18 nm allows to extend the polarization analysis to these very high harmonic orders.

The experimental setup using the multilayer mirror for the polarization analysis is shown schematically in Fig. 1. The gas jet (diameter about 1 mm) position is centered at the peak harmonic yield, i.e., about 0.5 mm in front of the laser focus. The radiation from the focus passes through a 1-μm aluminum filter that blocks the fundamental light and all the high harmonics beyond its L absorption edge, which is just beyond the 61st harmonic (H61). The transmitted light propagates to the Mo-Si multilayer mirror with an aperture of 14×24 mm^2. Finally, the reflected light is detected by a microchannel plate.

The calculated reflectivity of the multilayer mirror is shown in Fig. 2 [20]. The reflectivity of the mirror depends on the angle of incidence Θ and on the wavelength. The mirror can be used in a wavelength range from 11 to 22 nm. At a given angle of incidence, the reflectivity of the mirror peaks at the wavelength given by the dotted curve on the right-hand side scale of the figure. The peak in the wavelength dependence (not shown) has a characteristic width of (fwhm) about 15 Å. This is a key ingredient in our frequency selection: the central maximum in the mirror's reflectivity acts as a frequency bandpass, selecting only a few (three to four) harmonics. A further reduction of this bandpass is possible by shifting the reflection maximum of the mirror over the L-absorption edge of the Al filter. As a result we select only a single harmonic, primarily the 61st at $\lambda\approx17$ nm, with an admixture of roughly 20% of the 59th harmonic.

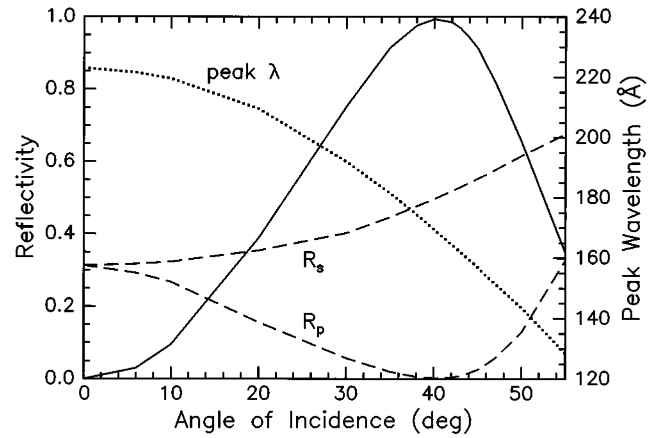

FIG. 2. Reflectivity properties of the multilayer mirror. Reflectivities R_s and R_p (dashed), analyzing power $(R_s-R_p)/(R_s+R_p)$ (solid), and peak wavelength (dotted, on right-hand scale).

Figure 2 also shows the polarization dependence of the reflectivity. It is seen that at an angle of $\Theta=40°$ the reflection of p-polarized light from the mirror, R_p, is practically zero. This implies that the multilayer mirror is an ideal polarizer and therefore more efficient than a grating or a metallic mirror in this spectral range. The analyzing power $(R_s-R_p)/(R_s+R_p)$ curve thus has its maximum at unity. By turning the multilayer mirror by a polar angle Φ about 360° around the HH beam in a plane vertical to the beam propagation it is possible to analyze the polarization state of the VUV radiation.

In analyzing the data we have to take into account that there is a residual signal from nonzero reflectivity at low-order harmonics. For wavelengths longer than 40 nm the reflectivity from the multilayer increases while the transmission from the Al filter is low but finite. Therefore, a difference measurement of two signals has been taken. First, we measured the total signal at $\Theta=37.5°$, and second we measured the ''background signal'' at $\Theta=42.5°$. This second signal was subtracted from the first value to yield the signal for the 61st harmonic only (with a small contribution from H59). The measurement for $\Theta=42.5°$ gives only the lower-order harmonic background since at that angle the maximum of the multilayer reflectivity is in the spectral region where the Al filter blocks the radiation completely.

III. RESULTS

We first show a typical result for the total photon yields. A characteristic HHG spectrum measured with the grating disperser setup and yielding absolutely calibrated photon numbers [11] is given in Fig. 3. The harmonic signal around the 61st order appears by roughly a factor of two higher compared to the lower-order plateau harmonics (< 41st order). This is caused by propagation (phase-matching) effects through the medium and depends sensitively on the focal

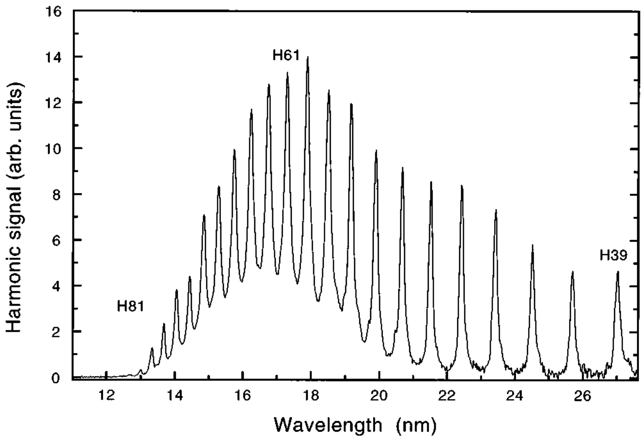

FIG. 3. HHG spectrum at $5(\pm 2)\times 10^{14}$ W/cm^2. The labels H39, H61, and H81 indicate harmonic orders.

geometry with respect to the gas jet [21]. The figure shows that the radiation from the 61st harmonic is optimized with respect to all other harmonics for the position of the gas jet (practically at the center of the laser focus) and that the harmonic is still in the plateau for our experimental conditions.

Inserting a quarter-wave plate into the laser beam in front of the lens produces pump pulses with different ellipticity values, allowing one to record the ellipticity dependence of the harmonic yield. The result for the 61st harmonic is presented in Fig. 4. We define the ellipticity ε as the ratio of the minor axis to the major axis of the electric field strengths. Each circle is the result from a single laser shot, the uncertainty resulting primarily from the laser pulse parameter variations. The data are compared to the prediction of lowest-order perturbation theory [13,22], $I_q \sim [(1-\varepsilon^2)/(1+\varepsilon^2)]^{q-1}$, where I_q is the intensity of the qth harmonic and ε the ellipticity of the laser. Obviously, lowest-order perturbation theory predicts a steeper falloff of the yield with ellipticity than the one we observe. This result is in agreement with previous work [13,20], where it was observed that the low-order harmonics follow the perturbative curve up to about $q \simeq 31$, while higher-order harmonics have a falloff ''frozen'' at about the value of $q=31$. In fact, the solid line in Fig. 4 for $q=35$ is the result of a least-squares fit to the experimental results. This is in accord with the theoretical single-atom response, using the 3D delta potential model. The average theoretical single-atom yield (smoothed out over the strong interference structures which appear in the plateau as a function of intensity) follows the perturbative law for harmonic orders up to about 31 and falls off with roughly the $q=31$ law for all higher orders.

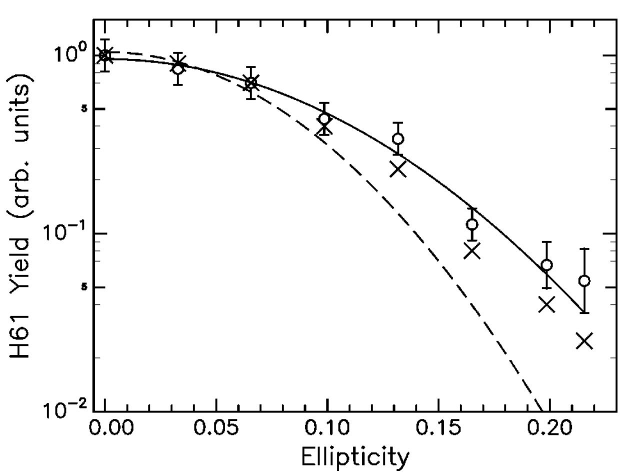

FIG. 4. Yield of the 61st harmonic from Ne vs laser ellipticity at a laser peak intensity of 10^{15} W/cm^2. Circles: present experiment. Uncertainties result from (i) laser fluctuations (20%) and (ii) detector background noise (10–20 %) at the largest ellipticities. Crosses: experiment of [5] (different wavelength and intensity), interpolated from graph, roughly similar uncertainties. Dashed line: Prediction of lowest-order perturbation theory $I_q \sim [(1-\varepsilon^2)/(1+\varepsilon^2)]^{q-1}$ with $q=61$. Solid line: Same dependence, but with $q=35$.

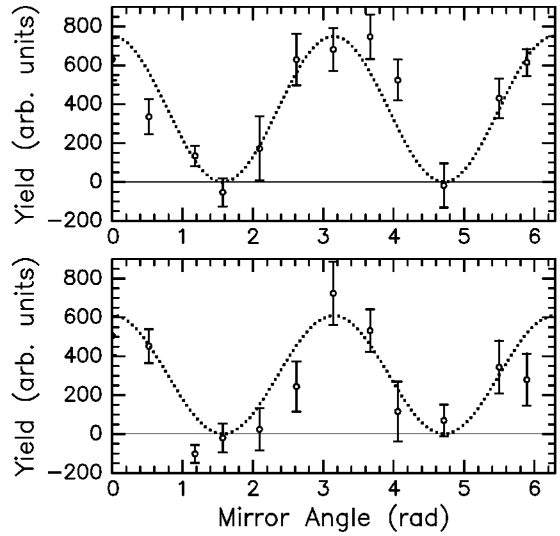

FIG. 5. Upper part: polarization of H61 for linearly polarized pump laser radiation (dotted line: theoretical $\cos^2$ curve). Lower part: polarization of H61 for slightly elliptically polarized pump laser radiation, $\varepsilon=0.03$ (dotted line: theory).

Using now the multilayer mirror [23], first we measured the harmonic polarization for linearly polarized driving laser radiation (upper plot in Fig. 5). The data points shown in Fig. 5 are the results of the difference between the $\Theta=37.5°$ and the $\Theta=42.5°$ measurements, leading to slightly negative values for some points. The error bars are given by the standard deviations resulting from the statistics of the 2×10 measured values at each Φ angle (10 shots for each of the two Θ values). The large statistical error in our measurement results from the low repetition rate of our laser, which allowed us to take only about 10 shots per Φ value, and a rather large fluctuation of the laser output energy, leading to an intensity spread in the measurements.

Theory yields linearly polarized harmonic radiation for linearly polarized driving field and thus a theoretical $\cos^2$ curve is superposed in the upper part of the figure. From a least-squares fit to the experimental data of the form $Y(\Phi) = Y_1 + Y_2\cos^2(\Phi+\varphi)$, we conclude that the ellipticity of the H61 radiation is $\varepsilon_{61} = \sqrt{|Y_1/(Y_1+Y_2)|} = 0.2 \pm 0.2$. The orientation of the major axis of the polarization ellipse of the harmonics with respect to the laser polarization direction is at $\varphi = -1° \pm 3°$. Thus, the experimental results are compatible with linear polarization of the H61 within experimental error, as expected.

In the lower plot of Fig. 5 the result from the measurement with slightly elliptical driving laser polarization $\varepsilon_{\mathrm{laser}} = 0.03 \pm 0.02$ is shown. Again there are some negative data points because of the difference measurement. We find that the ellipticity of the H61 is zero to within error: $\varepsilon_{61} = 0.0 \pm 0.3$. The rotation angle in this case is $\varphi = -0.3° \pm 3°$ [24].

In fact, as noted in [12], the measurement of the contrast ratio $Y_1/(Y_1+Y_2)$ leads only to an upper bound $|\varepsilon_{\mathrm{max}}|$ rather than to the harmonic ellipticity ε_{61} itself. The reason is the possibility of partial polarization of the harmonic light. The harmonic polarization varies in dependence of the pump pulse profile in space and in time through the intensity dependence of the qth component of the time-dependent dipole. This implies that the space- and time-averaged harmonic field, measured in the experiment, is only partially polarized.

Thus, for slightly elliptical laser polarization the polarization of the 61st harmonic remains linear and is oriented parallel to the incoming laser polarization. The dotted line gives the result from the theoretical simulation, which is in agreement with these findings. In fact, the difference from a $\cos^2$ curve is not visible in the figure. On the average, for the driving laser ellipticity of 0.03, the field components along the minor and major axes of the harmonic field emitted by an atom have a ratio that is even smaller, roughly 0.015. The integration of the propagation further reduces this ratio significantly. For the macroscopic response, the theoretical results give a much lower bound on the harmonic ellipticity, yielding evidently $\varphi = 0$ and $\varepsilon_{61} = 0$ for $\varepsilon_{\mathrm{laser}} = 0$. For $\varepsilon_{\mathrm{laser}} = 0.03$ the theory yields $\varphi = -0.2°$ and $\varepsilon_{61} < 0.001$. One has to bear in mind that the theoretical results have an uncertainty due to the uncertainty of the atomic response model: since the model is so successful in describing even the quantitative HHG yield, however, we presume this uncertainty is not relevant here.

We have also performed a measurement at $\varepsilon_{\mathrm{laser}} = 0.1$ ellipticity of the driving laser. However, our signal statistics was too low to allow the extraction of $\varepsilon_{\mathrm{max}}$ and φ with a reasonable error estimate.

We are planning to use the same analyzing arrangement to study a high harmonic from 800-nm driving laser radiation. The new Ti:Sa driving laser will give vastly improved statistics and thus much smaller error bars due to its better stability and much higher repetition rate.

IV. DISCUSSION AND CONCLUSIONS

In order to probe the HHG process in a ''complete'' experiment, both a variation of the driving force [the incoming laser beam(s)] and differential detection of the products [the harmonic radiation, the remaining atom or ion and the ejected electron(s)] must be performed. As a study of the angular dependence of the HHG, the driving laser polarization was made elliptical. In purely perturbative situations the emitted harmonic radiation is predicted to have the same polarization as the incoming light [22] for ground-state atoms, since the initial and final state are both known and unpolarized. In the present case, however, the atomic system can absorb and retain an arbitrary amount of (linear and angular) momentum, part of which can be carried away by ionized electrons. The polarization properties of the emitted light result from a transition from a well-defined initial state (ground-state atom and elliptically polarized incident laser radiation) to an unknown final state, the atomic part of which is not being detected. Therefore, the atomic harmonic response can in principle have any polarization the degree of which yields information on the physics of the HHG process. For low-order harmonics, where presumably atomic resonances play a predominant role, large variations of ellipticity of the harmonics have indeed been observed [13]. In the experiment, however, an average is taken over a large ensemble of atoms in the focus, which experience in general different laser intensities. The emitted harmonic light from the whole focal sample is therefore a superposition of many atomic responses, which tends to decrease the observed polarization, smoothing out its fluctuations.

We have performed a measurement of the polarization of a selected very-high-order harmonic of 1053-nm driving laser radiation, emitted in neon. The experiment used a multilayer mirror that presents a convenient polarizer in the XUV range with a near-perfect polarization analyzing power and a high reflectivity. We observe no rotation angle between harmonic polarization and laser polarization with increasing laser ellipticity for linear and slightly elliptical laser polarization. In both cases the polarization of the 61st harmonic is still linear within our experimental error. These results are in agreement with theoretical calculations [7] and measurements [14] for lower-order harmonics. In these previous experiments and calculations, in the cutoff region the harmonic ellipticity was much smaller than the fundamental ellipticity and a very small rotation angle of the harmonic polarization with respect to the fundamental one was observed for high degrees of ellipticity of the fundamental. On the other hand, for the harmonics in the plateau, it was reported that the polarization has a significant degree of ellipticity (however, smaller than the fundamental ellipticity) and the major axis of the harmonic polarization rotates by a large offset angle from the driving field polarization.

Our results extend the observed range to the 61st harmonic, for intensities such that it is just within the plateau region. In contrast to harmonics of order 35 and below, the total yield falloff with ellipticity is much slower than perturbative for these high harmonics, in agreement with [20]. From our polarization measurement we find that for a slightly elliptically polarized driving field there is no rotation of the harmonic ellipse with respect to the fundamental light. For linear and slightly elliptical laser polarization the 61st harmonic is linearly polarized. Therefore, if ellipticity switching is applied [25] to create fs harmonic pulses, one will obtain practically purely linearly polarized harmonic light when considering harmonics around order 61 (or higher).

Our theoretical results support the experimental findings and agree qualitatively with previous results [5]. The resulting single-atom harmonic yield around order 61 has an ellipticity that is comparable to but smaller than the fundamental laser ellipticity. Ellipticity and axis offset angle can exhibit larger values in the plateau. However, for these high harmonics, the harmonic yield has always minima at the intensities where ε and φ are significant. The propagated macroscopic response at a driving laser ellipticity of 0.03 has thus an ellipticity and offset angle that are much smaller than the present experimental uncertainty. In this sense the 61st harmonic behaves ''semiclassically'' in a most pronounced way, exposing none of the nonlinear polarization of the driving field.

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