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Generation of short-pulse tunable xuv radiation by high-order frequency mixing

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The generation of tunable short-pulse xuv radiation between 90 and 40 nm by nonlinear mixing of a fixed-frequency high-power Ti:sapphire laser with tunable radiation is described. As a tunable source an optical parametric generator pumped by part of the Ti:sapphire laser radiation has been used, producing tunable radiation in the range of about 520 to 650 nm with a peak power of up to 0.5 GW.

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For the generation of tunable coherent radiation in the xuv spectral range, nonlinear optical techniques have to be considered, as broadly tunable laser systems for this spectral range do not exist. In order to convert tunable visible radiation into the xuv, very-high-order processes are needed, which has become possible due to recent progress in nonperturbative high-order harmonic generation. Using intense short-pulse radiation mainly from Nd:glass [1], Ti:sapphire [2,3], or KrF excimer lasers [4], high-order harmonics have been generated with shortest wavelengths below 10 nm. Reference [5] gives an overview of the basic theory for high-order harmonic generation and some experiments.

Among the high-peak-power laser systems, only Ti:sapphire is broadly tunable and would therefore allow direct generation of tunable xuv radiation via the high-order harmonic process. However, this seems not to be practicable, as the high-peak-power systems are relatively complex and difficult to tune, due to the involved chirped pulse amplification scheme. Therefore, it seems to be more flexible to mix the radiation of a high-power laser (ω_P)with a less powerful but tunable light source (ω_{tunable}), generating tunable photons at $n\omega_P \pm k\omega_{\text{tunable}}$ ($n+k$ is odd).

A first high-order degenerate mixing experiment using ω_P and its second harmonic, $2\omega_P$, has been performed by Perry and Crane [6], and mixing of fixed-frequency high-peak-power KrF and ArF excimer laser radiation has been reported recently [7]. In this contribution we describe high-order frequency mixing experiments of fixed-frequency high-peak-power Ti:sapphire laser radiation at 813 nm with tunable radiation from an optical parametric generator (OPG) [8], allowing the generation of tunable short-pulse radiation in the xuv spectral range down to the highest harmonics observed so far. The OPG has been pumped by frequency-doubled radiation of part of the Ti:sapphire laser radiation and delivered peak powers of up to 0.5 GW, tunable in the range (so far) of 520 to 650 nm.

Figure 1 shows the experimental setup. The pump source for the experiments is a 150-fs Ti:sapphire laser (BMI ALPHA 10A) operating at 813 nm with an energy of up to 100 mJ at a repetition rate of 10 Hz. The principal setup is described in Ref. [9]. For the experiments, about 50 mJ have been used after clipping the output by a circular aperture. This radiation is sent through a 3-mm KDP crystal to generate about 8 mJ of second-harmonic radiation. The beam splitter BS1 separates the second harmonic from the fundamental. In the OPG about 10% of the pump beam (separated by beam splitter BS2) is focused with a telescope into a 5-mm BBO crystal (type I). The parametric fluorescence generated in this first pass is reflected back by mirror M1. The distance between M1 and the BBO crystal is about 40 cm. The reflected seed radiation is amplified in the second passage through the BBO crystal. For this, the beam size of the remaining pump beam is reduced with a single lens to a diameter of about 4 mm. The amplified pulse passes the beam splitter BS3.

The output beam of the OPG is increased by a telescope to roughly 2 cm and then via beam splitter BS5 combined with the remaining fundamental radiation (about 40 mJ). Both beams are focused by one 140-mm lens into the gas jet of a 1-mm-diam pulsed nozzle, close to the exit of the nozzle. As nonlinear media, noble gases have been used at backing pressures of about 2 bar, corresponding to densities in the gas jet of 10^{17}–10^{18} cm^{-3} at the interaction region. The generated radiation is analyzed with a Jobin-Yvon LHT 30 monochromator and detected with a microchannel-plate detector.

The OPG delivered (behind BS3) up to 350 μJ at 570 nm. The tuning range was 520–650 nm, which is less than the

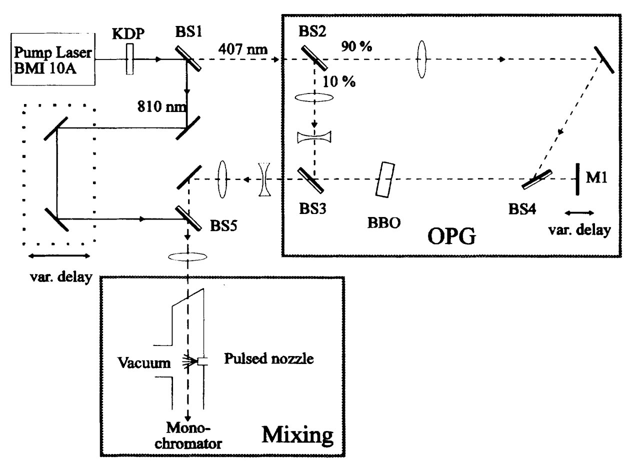

FIG. 1. Experimental setup. The radiation of the Ti:sapphire pump laser is frequency doubled. The second harmonic is separated and used to pump an optical parametric generator (OPG). The fundamental radiation and the tunable radiation from the OPG are mixed in the gas jet of the pulsed nozzle.

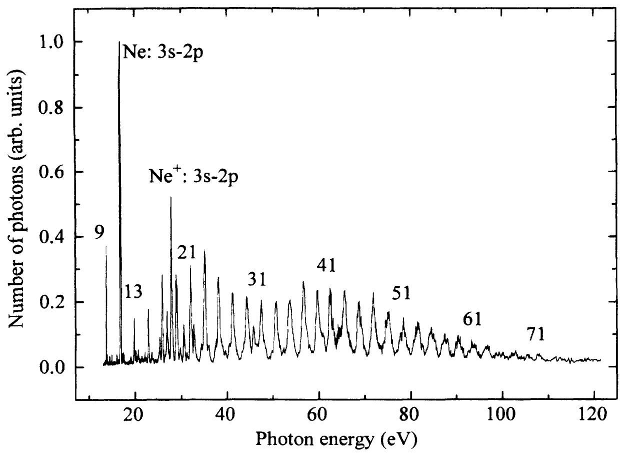

FIG. 2. Typical harmonic spectrum of the Ti:sapphire laser (at 813 nm) in neon (pump intensity $\approx 10^{16}$ W cm^{-2}).

theoretical tuning range of roughly 490–810 nm for the signal wave, and is probably caused by the limited transmission of mirror BS4. The pulse width of the OPG radiation was estimated by a cross correlation to be slightly less than 600 fs, which agrees with the theoretical group-velocity difference of pump and signal wave in BBO, which is 0.12 ps/mm at the wavelengths mentioned here. The time bandwidth product $\Delta t \Delta f$ was about 2. With a pulse duration of 600 fs the peak power of the OPG was above 0.5 GW.

With the described setup, harmonic generation and frequency mixing experiments have been performed with different noble gases. With the fundamental alone, harmonics up to the 71st order have been observed in neon (Fig. 2), with shortest wavelengths around 11.5 nm, limited by the efficiency of the monochromator grating. With argon, krypton, and xenon the maximum harmonic order decreased to 33 (24 nm), 31 (26 nm), and 19 (42 nm), respectively, which is in good agreement with estimations of the cutoff energy $I_{\rm ion}+\alpha U_P$, where $I_{\rm ion}$ is the ionization potential of the material and U_P the ponderomotive potential, which is proportional to the pump intensity. Assuming as maximum for U_P a value calculated with the saturation intensity due to optical

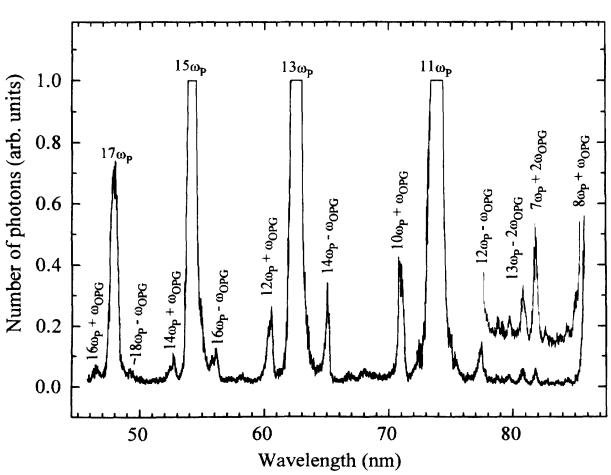

FIG. 3. Part of the mixing spectrum in xenon (λ_P 813 nm; I_P 10^{15} W cm^{-2}; $\lambda_{\rm OPG}$ 555 nm).

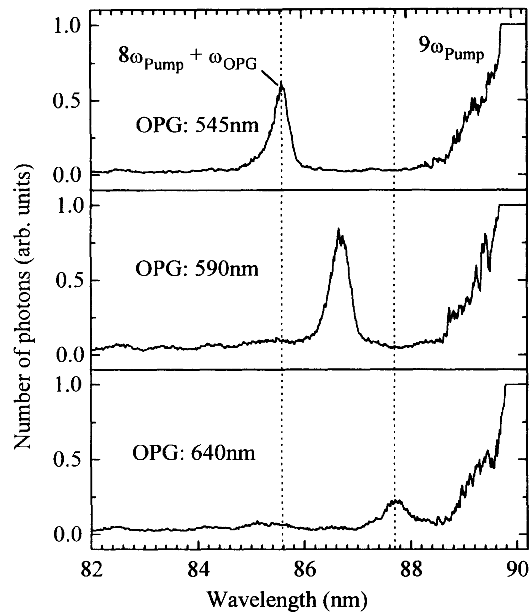

FIG. 4. Demonstration of the tunability of the $8\omega_P+\omega_{\rm OPG}$ mixing signal. The OPG has been tuned to three different wavelengths.

field ionization [10], α lies in the range of 2.1–2.3, in good agreement with Ref. [3]. Mixing signals could so far only be seen in argon, krypton, and xenon, with strongest signals in xenon, which has mostly been used in the experiments.

Figure 3 shows as an example a mixing spectrum for xenon in the range of 45–87 nm. In addition to the harmonics of the fundamental, all sum and difference frequency mixing signals $n\omega_P \pm \omega_{\rm OPG}$ (n even) can be seen down to 40 nm. Around 80 nm mixing signals with two OPG photons are also resolved at higher sensitivity. The intensity of the mixing signals (with one OPG photon) is at present between one and two orders smaller than the intensity of the harmonics (which are saturated in Fig. 3) and follow the general decrease of the harmonics towards shorter wavelengths. With a higher spectral resolution, Fig. 4 demonstrates as an example the tuning of one sum frequency mixing signal in the vicinity of the ninth harmonic. Sum mixing signals have been observed down to OPG energies of about 100 μJ, and up to the present maximum OPG energy of 350 μJ, the increase of the mixing signal has been almost linear. As energy and beam

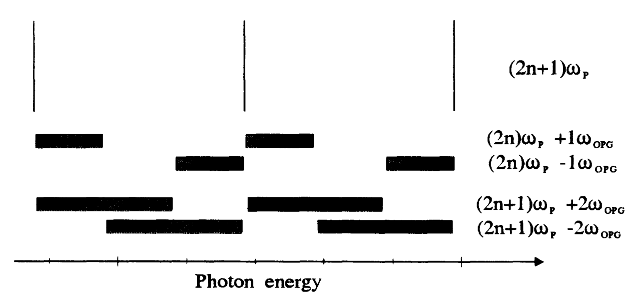

FIG. 5. Theoretical tuning range between subsequent odd harmonics $(2n+1)\omega_P$, $n=1,2,\ldots$, considering sum and difference frequency mixing processes with one and two OPG photons $\omega_{\rm OPG}$.

quality (focusability) of the OPG were not optimum in these experiments, it is expected that the mixing signals can be further strongly enhanced. For this a modified OPG setup with three passages of the generated radiation through the two BBO crystals will be realized.

With the present tuning range of the OPG and the one-OPG-photon mixing process, about 30% of the spectral range between two harmonics can be covered. This is extendable to more than 60%, if the OPG can be operated over the whole theoretical tuning range of about 490–810 nm. If also the weaker two-OPG-photon mixing processes are included, almost the whole spectral range between the harmonics can be covered, as illustrated by Fig. 5. Only just around the odd harmonics a small gap will remain, depending on how close the OPG is operated near degeneracy, where the bandwidth strongly increases.

With the described mixing scheme, tunable xuv radiation up to the highest harmonic may be generated. It is expected that at higher OPG intensities and with further optimizations the mixing signals can be strongly increased. In mixing experiments with two fixed-frequency excimer laser fields of similar intensity, fifth- and seventh-order mixing signals comparable to the corresponding harmonic signals have been observed [7]. Furthermore, the scheme may also be of interest for studies of the harmonic generation process with two independent frequencies and polarizations.

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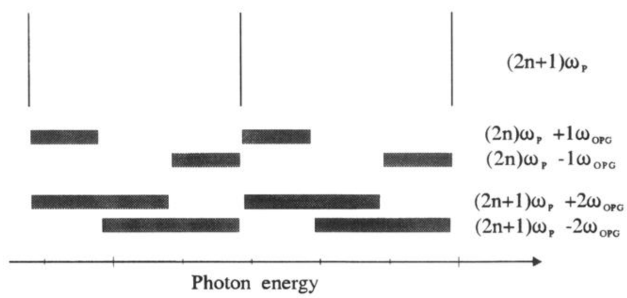

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