Generation of 10 \( \mu \text{W} \) relativistic surface high-harmonic radiation at a repetition rate of 10 Hz

J Bierbach\(^1\), C Rödel\(^{1,2}\), M Yeung\(^3\), B Dromey\(^3\), T Hahn\(^4\), A Galestian Pour\(^1\), S Fuchs\(^1\), A E Paz\(^{1,2}\), S Herzer\(^1\), S Kuschel\(^1\), O Jäckel\(^{1,2}\), M C Kaluza\(^{1,2}\), G Pretzler\(^4\), M Zepf\(^3\) and G G Paulus\(^{1,2,5}\)

\(^1\) Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany
\(^2\) Helmholtz-Institut Jena, Fröbelstieg 3, 07743 Jena, Germany
\(^3\) Centre for Plasma Physics, School of Mathematics and Physics, Queen’s University Belfast, BT7 1NN, UK
\(^4\) Institut für Laser- und Plasmaphysik, Heinrich-Heine-Universität Düsseldorf, Universitätsstraße 1, 40225 Düsseldorf, Germany

E-mail: gerhard.paulus@uni-jena.de

New Journal of Physics 14 (2012) 065005 (9pp)
Received 12 February 2012
Published 7 June 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/6/065005

Abstract. Experimental results on relativistic surface HHG at a repetition rate of 10 Hz are presented. Average powers in the 10 \( \mu \text{W} \) range are generated in the spectral range of 51 to 26 nm (24–48 eV). The surface harmonic radiation is produced by focusing the second-harmonic of a high-power laser onto a rotating glass surface to moderately relativistic intensities of \( 3 \times 10^{19} \text{ W cm}^{-2} \). The harmonic emission exhibits a divergence of 26 mrad. Together with absolute photon numbers recorded by a calibrated spectrometer, this allows for the determination of the extreme ultraviolet (XUV) yield. The pulse energies of individual harmonics are reaching up to the \( \mu \text{J} \) level, equivalent to an efficiency of \( 10^{-5} \). The capability of producing stable and intense high-harmonic radiation from relativistic surface plasmas may facilitate experiments on nonlinear ionization or the seeding of free-electron lasers.

\(^5\) Author to whom any correspondence should be addressed.
1. Introduction

The generation of high-order harmonics of high-power femtosecond laser pulses is the most prominent way to create extreme ultraviolet (XUV) coherent radiation and, at the same time, the shortest available pulse durations, i.e. attosecond pulses [1]. In the last decade, attosecond pulses produced by high-harmonic generation (HHG) in gases had a considerable impact on the development of experimental techniques in attosecond laser physics [2–4]. The efficiency of this HHG process is typically in the order of $10^{-6}–10^{-5}$ and has been the topic of a recent review [5]. However, many experimental approaches would benefit from higher photon fluxes or higher XUV pulse energies. Unfortunately, the process of HHG in gases cannot easily be scaled to higher XUV intensities and higher photon energies. Scaling the latter is largely limited by the ionization of the target gas [6], which defines an upper boundary for the intensity of the driving laser pulses. With increasingly powerful lasers, the photon flux can only be increased by using longer focal lengths. Applying this strategy, XUV pulses with energies of 1 µJ [7] up to 10 µJ [8] have been reported at a laser repetition rate of 10 Hz. This corresponds to a maximum average power of 100 µW [8]. An efficiency of HHG in gases of $10^{-4}$ and sub-µJ harmonic pulse energies have been reported using an advanced scheme that combines loose focusing and two-color laser fields [9].

The inherent limitations of HHG in gases do not exist for relativistic surface high-harmonic generation (SHHG) because a fully ionized relativistic plasma is used, i.e. the full potential of the highest achievable laser intensities can be exploited. The laser pulses are focused onto a surface to an amplitude of the normalized vector potential of $a_0^2 = (I \cdot \lambda^2)/(1.37 \times 10^{18} \text{W cm}^{-2} \cdot \text{µm}^2) \geq 1$, such that the surface electrons oscillate at relativistic velocities. Here, $\lambda$ is the laser wavelength and $I$ its intensity. For oblique incidence and $p$-polarization, the dominant process of energy transfer to the plasma occurs via the electric field component normal to the target surface. When the laser pulse is reflected at the relativistically oscillating plasma surface, its electromagnetic field is strongly modulated [10]. Accordingly, the reflected spectrum will contain high harmonics of the driving laser field. In the time domain, the harmonics correspond to a train of attosecond pulses. This simplified model of SHHG driven by a `relativistically oscillating mirror’ (ROM) [11] includes most of the essential physics. The spectral characteristics of the harmonic emission were predicted by an extended version of this model [12]. The efficiency of ROM harmonic generation in the ultra-relativistic limit $a_0 \gg 1$ follows the spectral power law $\eta \sim (\omega/\omega_0)^{-8/3}$, which predicts high efficiencies, e.g. $\eta = 10^{-4}$ for the 30th harmonic. Such efficiencies would enable attosecond XUV sources with pulse energies orders of magnitude higher than the current state-of-the-art. In fact, using
ultra-relativistic intensities it has been shown that bright harmonics up to keV photon energies can be generated [13].

Apart from delivering relativistic intensities to the target surface, also a well-defined plasma density gradient is essential for the SHHG process. In order to achieve efficient SHHG with a low divergence, the main pulse needs to interact with a steep plasma density gradient [14]. This can be realized by suppressing those prepulses that are intense enough to ionize the target before the arrival of the main pulse, which typically implies the use of contrast enhancement techniques such as plasma mirrors [15] or nonlinear optical filters, see, e.g. [16, 17]. Another practical issue for SHHG is the local destruction of the surface by each laser shot, such that a fresh surface of optical quality has to be provided for the next laser shot. For SHHG at nonrelativistic intensities, a rotating glass target with interferometric stabilization has been used at a repetition rate of 1 kHz [18]. However, relativistic SHHG, having more favorable properties like lower divergence [19] and higher photon energies [20], is harder to realize. As a consequence, previous experiments on ROM harmonic generation were performed in single-shot mode only.

We report on the demonstration of ROM harmonic emission performed at a repetition rate of 10 Hz using a computer-controlled rotating glass surface. A divergence of 26 mrad has been measured for the ROM harmonics emission in good agreement with previous results. The XUV emission was recorded using a calibrated spectrometer [21] thus enabling an absolute determination of the harmonics pulse energy and efficiency. We report on harmonic pulses with $\mu J$ energies and efficiencies of $10^{-7}$–$10^{-5}$ leading to an average power of >$10 \mu W$ for the observed XUV harmonic emission. Surface high-harmonic radiation is thus becoming a competitive source in terms of pulse energy.

2. Experimental setup

The experiment was carried out at Friedrich Schiller University Jena using the Ti:sapphire laser system ‘JETI-40’, which provides 30 fs pulses with an energy of 0.7 J at a 10 Hz repetition rate. Figure 1 displays the experimental setup. For reasons of contrast enhancement the laser pulses are frequency doubled using a 0.7 mm thick potassium dihydrogen phosphate (KDP) crystal. As SHHG requires horizontal polarization [10] and since type-I phase matching is used for second-harmonic generation (SHG), the polarization of the fundamental laser pulse is first rotated to vertical orientation using a half-wave plate. In order to optimize the horizontal alignment of the polarization and the pulse energy of the second harmonic, the angles of the wave plate and the KDP crystal are fine-tuned. To this end the polarization of the generated 400 nm pulses is adjusted to horizontal orientation by aligning the crystal axis and then SHG is optimized in terms of conversion efficiency by fine-tuning the fundamental’s polarization using the half-wave plate.

Two multilayer mirrors, which are highly reflective for 400 nm and anti-reflective for 800 nm, are used in order to suppress the fundamental (800 nm) pulse intensity by more than a factor of 10$^5$. The 400 nm pulses are focused onto a fused silica target ($<1$ nm root mean square (RMS) roughness) at 45° angle of incidence in $p$-polarization by an $f/2$ off-axis aluminum parabola. By imaging the focus of the frequency doubled pulses using a microscope objective (see figure 1(b)), it is found that 29% of the entire pulse energy of up to 100 mJ is contained in the focal area of 3.6 $\mu m^2$ (enclosed by the contour line of the full-width at half maximum (FWHM)). Based on the analysis in [22], where the preservation of the pulse duration for transform limited pulses in thin crystals has been shown, the pulse duration of the second
Figure 1. (a) Experimental setup for relativistic SHHG using the second harmonic of 800 nm, 30 fs laser pulses: The polarization of the incident laser beam is rotated such that the resulting 400 nm, 30 fs pulses are focused to relativistic intensities onto a rotating glass target at 45° incidence angle in p-polarization. Dichroic mirrors suppress the fundamental pulse intensity by more than a factor of $10^5$. (b) Intensity distribution of the second harmonic at the focus position: The FWHM contour line encloses an area of 3.6 $\mu$m$^2$ and contains 29% of the total second harmonic pulse energy of 100 mJ resulting in a relativistic intensity of $\sim 3 \times 10^{19}$ W cm$^{-2}$. (c) Setup of the calibrated XUV spectrometer [21] that is used for the photon flux measurement.

The harmonic emission is measured in the specular direction using two different spectrometer setups. A flat-field grating spectrometer is used to determine the divergence of the harmonic beam (figure 3). In this configuration, the incident XUV radiation is dispersed using a 1200 lines mm$^{-1}$ grating [24] and detected by an XUV camera (back-thinned
Figure 2. (a) The second-harmonic pulse energy is measured as a function of the fundamental’s pulse energy. The error bars indicate the standard deviation of sets of 5 shots. For low input energy, a quadratic scaling for the conversion is assumed (dashed line). For high energies the efficiency is found to saturate at $\sim 18\%$. (b) Pulse contrast of the 800 nm laser pulses measured by a third-order autocorrelator (black line). The contrast of the 400 nm pulse (red line) is calculated using the measured energy dependence of the SHG process, see panel (a).

Figure 3. An approximately uniform divergence of $\sim 26$ mrad (red line) is measured for different ROM harmonic orders (black dots).

Andor DO940N). In the horizontal plane, the XUV emission is recorded with an acceptance angle of 12 mrad. Therefore, the divergence can only be estimated based on the fraction of the harmonic beam that is collected by the spectrometer’s aperture [19]. A larger angular range of $\sim 50$ mrad is measured by scanning the harmonic beam over the aperture. This is realized by translating the focusing parabola and thus tilting and translating the centroid beam with respect to the optical axis of the spectrometer. The angular distributions of the 12th to 15th harmonics show a uniform divergence of $\sim 26$ mrad (figure 3), which is a consequence of the spatial denting of the plasma surface due to the ponderomotive force [25].

In a second spectrometer setup, an XUV spectrometer (figure 1(c)), which was previously calibrated with respect to the incident photon flux [21], was used. Two 0.2 $\mu$m aluminum foils located at the entrance aperture of the spectrometer block the intense visible radiation. The transmitted XUV emission [26] is imaged by a nickel-coated toroidal mirror onto the CCD camera. A transmission grating consisting of freestanding gold bars with 1000 lines mm$^{-1}$ is...
Figure 4. (a) XUV spectra generated by 250 subsequent laser shots at a repetition rate of 10 Hz. A false color scale is used for the number of counts. (b) Averaged spectral energy per pulse and average power of the surface harmonics in comparison with the spectrum from a single shot under optimized conditions.

used to disperse the XUV radiation. The harmonics’ energy is determined by taking into account the filter transmission, the absolute sensitivity of the spectrometer [21] and the fraction of the surface harmonics that enters the spectrometer aperture.

3. Experimental results

We recorded SHHG spectra for every laser pulse at a repetition rate of 10 Hz. 250 subsequent spectra taken over a period of 25 s are displayed in figure 4(a). Each line represents a single spectrum reaching from the 8th up to 15th harmonic of the 400 nm driver laser field. It should be noted that harmonics with frequencies lower than the plasma frequency can also be produced by the nonrelativistic mechanism of coherent wake emission (CWE) [27]. For the fused silica targets used, the maximum plasma frequency is approximately situated at the 10th harmonic and constitutes the cutoff frequency for CWE harmonics [28]. Consequently, higher harmonic orders must be produced by the ROM process. The fact that there is no significant change in divergence or efficiency around the 10th harmonic order suggests that the ROM is the dominant process for all the harmonics observed at our conditions. Nevertheless, the CWE mechanism might contribute to the 8th and 9th harmonic orders as we also recorded these orders at non-relativistic intensities. In fact, the efficiency of $10^{-5}$ (table 1) conforms to the discussion given in [27].

The spectral energy distribution averaged over all 250 spectra consists of harmonic lines and a broad XUV background signal as shown in figure 4(b). It is worth mentioning that the double-peak structure in the harmonic spectra is evidence of a spectral modulation owing to the generation of unequally spaced attosecond pulses [23]. This effect arises from the superposition of the ROM process and a temporal denting of the plasma surface due to the radiation pressure at relativistic intensities.

We have calculated the stability of ROM harmonic generation based on the data set of 250 spectra. A relative deviation of $\sim 30\%$ for the SHHG intensity and $<10\%$ for the background...
An analysis of the 250 spectra presented in figure 4 reveals pulse energies of \( \mu J \) for individual harmonic lines thus yielding an efficiency of \( 10^{-5} \) and an average power of the order of \( 10 \mu W \). The analysis of a ROM spectrum from single shot operation under optimized conditions shows a difference of half an order of magnitude.

| \( \Omega/\omega_{\text{SHG}} \) | \( E_\nu (\mu J) \) | \( \eta \) | \( P_\nu (\mu W) \) | \( E_\nu (\mu J) \) | \( \eta \) | \( P_\nu (\mu W) \) |
|-------|-------------|-------|-------------|-----------------|-------|-------------|
| 8     | 0.6         | 1.6 \( \times \) \( 10^{-5} \) | 6.0           | 1.2             | 3.3 \( \times \) \( 10^{-5} \) | 12.0    |
| 9     | 0.4         | 1.1 \( \times \) \( 10^{-5} \) | 4.3           | 0.9             | 2.5 \( \times \) \( 10^{-5} \) | 9.4     |
| 10    | 9.7 \( \times \) \( 10^{-2} \) | 2.5 \( \times \) \( 10^{-6} \) | 1.0           | 0.3             | 7.1 \( \times \) \( 10^{-6} \) | 2.7     |
| 11    | 6.3 \( \times \) \( 10^{-2} \) | 1.7 \( \times \) \( 10^{-6} \) | 0.6           | 0.2             | 5.7 \( \times \) \( 10^{-6} \) | 2.2     |
| 12    | 8.1 \( \times \) \( 10^{-2} \) | 2.1 \( \times \) \( 10^{-7} \) | 8.1 \( \times \) \( 10^{-2} \) | 8.4 \( \times \) \( 10^{-2} \) | 2.2 \( \times \) \( 10^{-6} \) | 0.8     |
| 13    | 6.6 \( \times \) \( 10^{-3} \) | 1.7 \( \times \) \( 10^{-7} \) | 6.6 \( \times \) \( 10^{-2} \) | 2.6 \( \times \) \( 10^{-2} \) | 7.0 \( \times \) \( 10^{-7} \) | 0.3     |
| 14    | 3.4 \( \times \) \( 10^{-3} \) | 9.0 \( \times \) \( 10^{-8} \) | 3.4 \( \times \) \( 10^{-2} \) | 1.5 \( \times \) \( 10^{-2} \) | 3.9 \( \times \) \( 10^{-7} \) | 0.2     |
| 15    | 3.4 \( \times \) \( 10^{-3} \) | 9.0 \( \times \) \( 10^{-8} \) | 3.4 \( \times \) \( 10^{-2} \) | 9.2 \( \times \) \( 10^{-3} \) | 2.4 \( \times \) \( 10^{-7} \) | 9.2 \( \times \) \( 10^{-2} \) |

was found. The fluctuations are mainly due to variations in the laser intensity and a slight drift of the target surface, caused by the vertical translation of the target. The latter was corrected manually at shot number 150 by repositioning of the target during the running measurement. Monitoring and positioning the target surface in a closed loop will lead to a substantial increase in stability.

The determination of SHHGG efficiency requires subtraction of the background XUV spectrum. We measure a total XUV background yield of 0.4 \( \mu J \) in the spectral range covered by the spectrometer. Integration of individual harmonics of the background-free spectra yields harmonic energies up to microjoules. A detailed list is given in table 1. It should be noted that for the computation of the efficiencies only that fraction of energy of the driving 400 nm pulse is taken into account that is focused to a normalized vector potential of \( a_0 \geq 1 \) and thus can be considered to be relevant for the ROM mechanism. For the measured focal intensity distribution shown in figure 1(b), 39 mJ of the frequency doubled pulse energy fulfills this condition. The energies for the harmonics in the spectral range of 24–48 eV result in a total average power of 12.3 \( \mu W \) for the observed ROM harmonic emission. The efficiencies at our conditions are of the order of \( 10^{-5}–10^{-7} \). Selected laser shots from single shot operation, however, produce considerably higher pulse energies and efficiencies, cf figure 4(b) and table 1. An obvious conclusion is that there exists a significant potential for further optimization. Nevertheless, it has to be admitted that the efficiency of ROM harmonics under the present conditions falls behind expectations created by theoretical results valid in the ultra-relativistic limit. Apparently, the ROM is only at the onset of being driven efficiently to the relativistic regime.

4. Conclusion

In conclusion, we demonstrate the first consecutively measured relativistic surface HHG at a repetition rate of 10 Hz. This establishes a compact source of high-intensity XUV pulses from

---

6 This is done in order to evaluate the ROM process itself. The efficiencies with respect to the entire SHG pulse energy of 100 mJ would be smaller by a factor of 0.39.
relativistic laser plasma interaction with a repetition rate that is suitable, e.g., for experiments on nonlinear photoionization. The ROM harmonic source has been characterized with respect to the beam divergence, XUV pulse energy and efficiency. At the moderately relativistic intensities used in this experiment, the ROM harmonic emission is at the brink of being capable of competing with HHG from gaseous media in terms of efficiency. There is, however, quite some potential for improvements. In the presented setup the plasma scale length is estimated by the SHG pulse contrast. In order to control and optimize the scale length with respect to harmonic energy, prepulses and different contrast enhancement techniques could be applied. Furthermore, the focal spot could be significantly improved using adaptive optics or a KDP crystal with a higher optical quality. The latter is also capable of increasing the conversion efficiency and energy of the SHG pulses.

Acknowledgments

This work was supported in part by the Deutsche Forschungsgemeinschaft via project TR 18 and by Laserlab Europe. We thank F Ronneberger and B Beleites for operating the JETI laser. CR acknowledges support from the Carl Zeiss Stiftung.

References

[1] Drescher M, Hentschel M, Kienberger R, Uberacker M, Yakovlev V S, Scrinzi A, Westerwalbesloh Th, Kleineberg U, Heinzmann U and Krausz F 2002 Nature 419 803–7
[2] Goulielmakis E et al 2010 Nature 466 739–43
[3] Cavaliere A L et al 2007 Nature 449 1029–32
[4] Pfeifer T, Abel M J, Nagel P M, Jullien A, Loh Z-H, Bell M J, Neumark D M and Leone S R 2008 Chem. Phys. Lett. 463 11–24
[5] Sansone G, Poletto L and Nisoli M 2011 Nature Photonics 5 655–63
[6] Colosimo P et al 2008 Nature Phys. 4 386–389
[7] Hergott J-F, Kovacev M, Merdji H, Hubert C, Mairesse Y, Jean E, Breger P, Agostini P, Carré B and Salières P 2002 Phys. Rev. A 66 021801
[8] Takahashi E, Nabetake Y and Midorikawa K 2002 Opt. Lett. 27 1920–22
[9] Kim I J, Lee G H, Park S B, Lee Y S, Kim T K and Nam C H 2008 Appl. Phys. Lett. 92 021125
[10] Lichters R, Meyer-ter-Vehn J and Pukhov A 1996 Phys. Plasmas 3 3425–37
[11] Gordienko S, Pukhov A, Shorokhov O and Baeva T 2004 Phys. Rev. Lett. 93 115002
[12] Baeva T, Gordienko S and Pukhov A 2006 Phys. Rev. E 74 046404
[13] Dromey B et al 2007 Phys. Rev. Lett. 99 085001
[14] Zepf M et al 1998 Phys. Rev. E 58 R5253
[15] Rödel C, Heyer M, Behmke M, Kübel M, Jäckel O, Ziegler W, Ehrt D, Kaluza M C and Paulus G G 2011 Appl. Phys. B 103 295
[16] Marcinkevicius A, Tommasini R, Tsakiris G D, Witte K J, Gaiauskas E and Teubner U 2004 Appl. Phys. B 79 547
[17] Jullien A et al 2005 Opt. Lett. 30 920–22
[18] Borot A et al 2011 Opt. Lett. 36 1461–3
[19] Dromey B et al 2009 Nature Phys. 5 146–52
[20] Thauery C et al 2007 Nature Phys. 3 424–9
[21] Fuchs S, Rödel C, Paz A E, Kuschel S, Wünsche M, Hilbert V, Zastrau U, Ziegler W, Förster E and Paulus G G 2012 Rev. Sci. Instrum. submitted
[22] Sidick E, Knoesen A and Dienes A 1995 *J. Opt. Soc. Am.* B **12** 1704–12
[23] Behmke M *et al* 2011 *Phys. Rev. Lett.* **106** 185002
[24] Kita T, Harada T, Nakano N and Kuroda H 1983 *Appl. Opt.* **22** 512–3
[25] an der Brügge D and Pukhov A 2007 *Phys. Plasmas* **14** 093104
[26] Hemmers D, Benzid M and Pretzler G 2012 *Appl. Phys.* B submitted
[27] Quere F, Thaury C, Monot P, Dobosz S, Martin P, Géindre J-P and Audebert P 2006 *Phys. Rev. Lett.* **96** 125004
[28] Tarasevitch A, Lobov K, Wünsche C and von der Linde D 2007 *Phys. Rev. Lett.* **98** 103902