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Pulsewidth-switchable ultrafast source at 114 nm

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Femtosecond laser sources with high repetition rate are fundamental tools enabling tabletop time-resolved and angle-resolved photoemission spectroscopy in solids. The UV and vacuum UV laser light required for the photoemission process can be generated through various frequency up-conversion techniques. We describe a VUV source at 114 nm (10.8 eV) based on an industrial grade ytterbium-doped ultrafast laser, a nonlinear pulsewidth selection stage, and two cascaded frequency tripling stages, first in crystals, second in xenon. The role of ionization in gas-based perturbative third harmonic generation phase-matching is analyzed using a simple theory, numerical simulations, and experimental data. The source features high photon flux, high repetition rate and adjustable time resolutions. Thereby, in combination with a state-of-the-art ARPES apparatus it enables the study of the electronic dynamics of the whole Brillouin zone in a large number of materials. ©

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The rapid expansion of femtosecond laser sources with high repetition rate considerably increases the performance of a number of spectroscopy techniques, including tabletop time-resolved and angle-resolved photoemission spectroscopy (tr-ARPES) in solids. tr-ARPES is an instrumental technique in condensed matter physics which enables access to the band structure of solids as well as exploring the dynamics of quasiparticles in quantum materials [1]. The characteristics of the light source used to generate photoelectrons largely determine the performances of any tr-ARPES experiment. For instance, the achievable time and momentum/energy resolutions are related to the optical pulsewidth and bandwidth, the achievable signal to noise ratio and acquisition time are related to the repetition rate and photon flux of the source, and the accessible momentum space is determined by its central wavelength. To efficiently probe the materials, vacuum ultraviolet (VUV) or extreme-ultraviolet (EUV) ultrafast sources are required. UV light at energies around 6 eV overcomes the work function of solids and is extensively used as probe in tr-ARPES [2] but it can detect only electronic

states in the proximity of the Gamma point. In order to access the whole Brillouin zone, higher energy photons are necessary.

Besides synchrotron and free-electron laser facilities, with limited access, tabletop sources based on frequency up-conversion of ultrafast lasers in the near infrared have been developed for this purpose. Depending on the up-conversion mechanism, these can be classified in two categories. First, some sources are based on non-perturbative high-harmonic generation (HHG) [3, 4], which allows access to the EUV range in a single step, often with limited photon flux and spectral resolution. Second, multiple stages of perturbative nonlinear processes can be performed to access the VUV range [5, 6], first in crystals for wavelengths above ~ 200 nm, then in gases. The technology shift from titanium-doped sapphire lasers to ytterbium-based lasers has allowed an increase in repetition rate of these tabletop VUV sources and triggered renewed interest to optimize them for tr-ARPES. In particular, cascaded frequency tripling of high power laser systems at a central wavelength of ~ 1 μ m has been investigated [7–10]. In these sources a first stage of third-harmonic generation (THG) is performed using second-harmonic generation (SHG) and sum-frequency generation (SFG) in nonlinear crystals. This is followed by a second THG stage in a rare gas or gas mixture which is a third-order perturbative nonlinear effect subject to phase matching (PM).

Here, we describe a high repetition rate VUV source based on cascaded frequency tripling of Fourier transform-limited (FTL) pulses with a duration easily switchable between 100 fs, 500 fs, and 3 ps. The 500 fs pulses delivered by the driving ytterbium-based laser at 1030 nm and 100 kHz can be used directly or sent to a nonlinear multipass cell (MPC) that performs either spectral broadening or spectral compression, depending on the input chirp. The pulses are then sent to a first crystal-based THG stage, then focused in a xenon cell to generate the ninth harmonic.

This unique setup allows us to observe and analyze the dependence of PM pressure on experimental parameters such as pulsewidth, intensity, and focusing conditions. In particular, although THG in gases has been observed and used for decades [11], the effect of ionization on the process phase-matching has not been considered in detail, in contrast with HHG. We provide such an analysis and show that it matches the experimentally observed THG efficiency as a function of gas pressure and pulsewidth. The versatility of this source at 114 nm (10.8 eV)

in terms of spectral/temporal width, combined with a photon flux in the 10^{12} ph/s range makes it very appealing for various tr-ARPES experiments.

We first analyze the influence of ionization in the PM of perturbative gas-based THG. Early analyses of this problem [11–13] establish the possibility to obtain PM conditions by compensating neutral gas dispersion by the Gouy phase. To include analytically the effect of ionization, we turn to a description localized in one plane, similar to what is done in HHG [14, 15]. In the following, we assume that this plane corresponds to the driving pulse waist. In contrast to HHG, in the perturbative THG case, the atomic dipole phase is not relevant. Including contributions from neutral atoms, self- and cross-phase modulation (SPM, XPM) induced by the fundamental, Gouy phase, and free electrons, the phase mismatch $\Delta k = k_{3\omega} - 3k_{\omega}$ can be written as

$$\Delta k = \frac{\Omega}{c} [(n_{3\omega} - n_{\omega}) + (n_{2X} - n_{2S}) I_{\omega}] + \frac{2}{z_R} + \frac{4e^2 n_e}{\Omega m_e \epsilon_0 c}, \quad (1)$$

where $\Omega = 3\omega$ is the angular frequency of the third harmonic, n_{ω} and $n_{3\omega}$ are the refractive indices at the fundamental and third harmonic wavelengths, c is the speed of light, z_R is the Rayleigh range, I_{ω} is the intensity of the fundamental, n_{2X} is the nonlinear index associated with XPM from the fundamental onto the third harmonic, n_{2S} is the nonlinear index governing SPM of the fundamental, e , m_e , and n_e are the electron charge, mass, and density, and ϵ_0 is the vacuum permittivity. At 343 nm, for intensities below 5×10^{13} W/cm² in xenon, ionization is well approximated by a three-photon process. Assuming a low ionization fraction and a Gaussian pulse profile, the electron density at the peak of the pulse is given by [16]

$$n_e = \frac{1}{4} n_0 \sigma_3 I_{\omega}^3 \sqrt{\frac{\pi}{3 \ln 2}} \Delta t, \quad (2)$$

where n_0 is the initial atomic density, and Δt is the Full Width at Half Maximum (FWHM) pulsewidth. The three-photon ionization cross-section of xenon $\sigma_3 \approx 1.2 \times 10^{-39} (\text{W}\cdot\text{m}^{-2})^{-3} \cdot \text{s}^{-1}$ is found by fitting ionization rates calculated using the method described in [17] in the multiphoton regime. Substituting this expression in Eq. (1), and removing the SPM/XPM term, which is found to be negligible in our conditions (two orders of magnitude lower than the neutral atoms contribution for $I_{\omega} = 10^{13}$ W.cm⁻²), the phase mismatch can be written as

$$\Delta k = \frac{\Omega}{c} p_{Xe} \Delta N_{Xe} + \frac{2}{z_R} + \kappa p_{Xe} \Delta t I^3 \quad (3)$$

$$\kappa = \frac{e^2 n_L \sigma_3}{\Omega m_e \epsilon_0 c} \sqrt{\frac{\pi}{3 \ln 2}}$$

where n_L is the Loschmidt constant, p_{Xe} is the xenon pressure, the refractive indices have been written as $n_{\omega,3\omega} = 1 + p_{Xe} N_{\omega,3\omega}$ to bring forward their linear pressure dependence, and $\Delta N_{Xe} = N_{3\omega} - N_{\omega} \approx -3.7 \times 10^{-4} \text{ bar}^{-1}$. Setting $\Delta k = 0$ allows us to find a relationship between the PM pressure and the intensity

$$I_{\omega}^3 = -\frac{1}{\kappa \Delta t} \left(\frac{\Omega}{c} \Delta N_{Xe} + \frac{2}{z_R p_{Xe}} \right). \quad (4)$$

The first term in the parenthesis, corresponding to the neutral dispersion, is negative, while the second one, corresponding to the Gouy phase, is positive. Figure 1 illustrates this relationship at a driving wavelength of 343 nm in xenon for three focusing conditions and pulse durations. When ionization is negligible

($I \rightarrow 0$), we recover the balance between neutral dispersion and Gouy phase, defining a first horizontal asymptote.

$$z_R p_{Xe} = -2 \frac{c}{\Omega \Delta N_{Xe}}. \quad (5)$$

As the intensity increases, the PM pressure increases because the neutral atoms dispersion must compensate for the ionization contribution. At high intensity the neutral atoms dispersion contribution must be much higher than the Gouy phase contribution to balance ionization, leading to a second vertical asymptote

$$I_{\omega}^3 = -\frac{1}{\kappa \Delta t} \frac{\Omega}{c} \Delta N_{Xe}. \quad (6)$$

The horizontal asymptote moves up to a higher pressure when the waist radius is decreased, and the vertical asymptote is shifted to a higher intensity when reducing the pulse duration. As in the case of HHG [15], shorter pulsewidths allow the possibility of phase-matching over a larger intensity range, because the peak intensity can be reached for a lower ionization fraction.

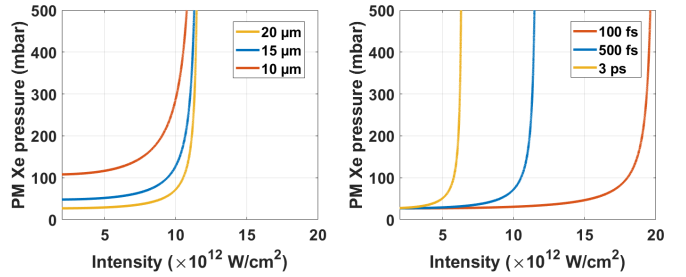


Fig. 1. PM pressure as a function of intensity of the driving pulse at 343 nm in the Xe cell. Left: different waist radii at fixed 500 fs pulse duration. Right: different pulse durations at fixed waist radius of 10 μm .

To capture physical effects beyond this simplified analysis, that considers phase-matching in a single plane at maximum intensity in space and time, we compare the experimental results with numerical simulations. These simulations are based on solving two coupled envelope equations for the fundamental and THG fields in (2+1)D geometry (r, z, t), assuming radial symmetry. They take into account diffraction, dispersion through a Sellmeier equation for xenon [13], THG, SPM, XPM, assuming a single $\chi^{(3)}$ value corresponding to a nonlinear index $n_2 = 5.2 \times 10^{-23} \text{ m}^2/\text{W}$ for all these processes, and ionization-related effects including free electron dispersion. This model allows to get a more accurate picture, taking into account the temporal, transverse and longitudinal spatial variations in intensity and phase for both beams. For comparison with experiments, the only slightly adjusted parameter is the waist radius, because it was difficult to estimate its precise value due to the limited resolution of the camera used.

We now turn to the description of the experimental setup, shown in Fig. 2. It starts with an Yb-doped industrial laser source (Tangor, Amplitude) that delivers 500 fs 300 μJ pulses at 100 kHz repetition rate and 1030 nm wavelength. This laser is delivered with a remote-controlled grating-based compressor unit that allows easy chirp adjustment of the output pulses. Using a set of flip mirrors, the pulses can either be sent directly to the crystal-based frequency tripling stage to seed the 500 fs beam-line, or to a MPC to manipulate the pulse duration. This MPC is

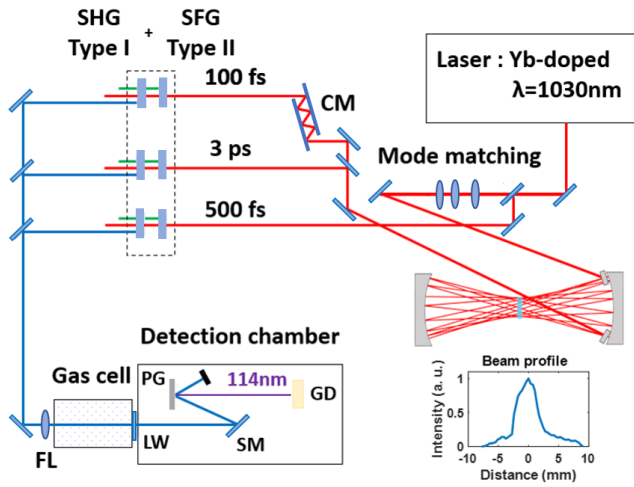


Fig. 2. GD: gold detector, SM: spherical mirror, PG: plane grating, LW: LiF window, FL: focusing lens, CM: chirped mirrors. Layout of the experimental setup. Inset: beam profile of the 114 nm pulses based on a knife edge measurement after the grating.

143 designed so that, if the input pulses are FTL, it compresses them
 144 down to 100 fs [18, 19], while if they are negatively stretched
 145 to 3 ps, it compresses the spectrum down to the FTL limit [20].
 146 Indeed, applying self-phase modulation on negatively chirped
 147 pulses results in instantaneous frequency shifts that suppress
 148 the edges of the input spectrum. This results in a very easy
 149 switch between 100 fs (temporal compression), 500 fs (initial
 150 laser source), and 3 ps (spectral compression) FTL pulses.

151 The MPC is composed of two concave mirrors with a radius
 152 of curvature of 300 mm separated by 550 mm in a Herriott con-
 153 figuration [21] inside which a 5 mm-thick AR-coated fused silica
 154 plate is inserted to provide nonlinearity [18]. An arrangement
 155 of three lenses is used to match the input beam to the stationary
 156 beam of the MPC, and the pulses are propagating through 15
 157 roundtrips before being sent out. At the output of the MPC,
 158 another flip mirror is used to send the pulses to a set of chirped
 159 mirrors introducing a total group-delay dispersion of -16000
 160 fs^2 (spectrally broadened pulses) or directly to the frequency
 161 tripling stage (spectrally compressed pulses). The spectral and
 162 temporal profiles of each beamline are shown in Fig. 3. The
 163 time-bandwidth products are 0.51, 0.43, and 0.5 for the 100 fs,
 500 fs, and 3 ps pulses respectively.

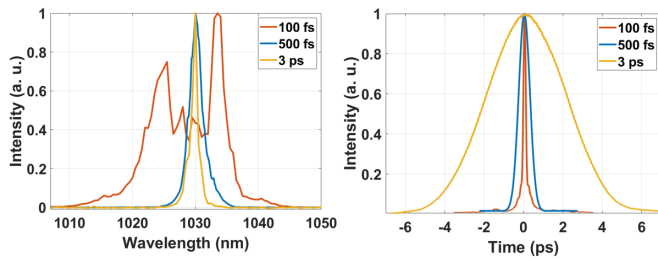


Fig. 3. Left: measured spectra at 1030 nm. Right: measured autocorrelation traces at 1030 nm.

164 The nonlinear crystals used in the first THG stage (all pro-
 165 vided by Crylight) must be adapted to the three different pulse
 166

167 durations. Each beamline of 100 fs, 500 fs and 3 ps with energies
 168 of 150 μJ , 300 μJ and 200 μJ respectively, is therefore sent to three
 169 parallel tripling stages. Each stage is based on SHG in a first
 170 BBO crystal (thicknesses of 0.5, 1, and 2 mm for 100 fs, 500 fs
 171 and 3 ps respectively), followed by SFG in a second BBO crystal
 172 (thicknesses of 0.75, 1.5, and 2 mm for 100 fs, 500 fs and 3 ps
 173 respectively). Before the second, gas-based THG stage, the avail-
 174 able energies at 343 nm are 15 μJ , 30 μJ , and 30 μJ for driving
 175 pulse durations of 100 fs, 500 fs, and 3 ps respectively. The lower
 176 pulse energy available at 100 fs duration is due to the losses
 177 of the chirped mirrors, and lower tripling efficiency. Although
 178 the pulse durations at 343 nm are not measured directly, the
 179 measured spectra indicate that the pulse durations at 1030 nm
 180 are approximately conserved at 343 nm.

181 The second THG stage consists of a xenon-filled cell in which
 182 the beam at 343 nm is focused, yielding a beam at 114 nm. Both
 183 beams enter the under-vacuum detection chamber transmitted
 184 through a lithium fluoride (LiF) window. The photon flux is
 185 estimated by measuring the drain current from the surface of a
 186 gold target [22] inserted just after the LiF window. Because the
 187 work function of gold is 5.2 eV while the photon energies of the
 188 343 nm and 114 nm light are 3.6 eV and 10.8 eV respectively,
 189 the detection of stray light is efficiently suppressed. We have also
 190 experimentally verified that illuminating the gold surface with
 191 the full power at 343 nm in the absence of THG (gas cell under
 192 vacuum), in the three different pulse duration cases, resulted in
 193 negligible measured current. A knife edge measurement of the
 194 114 nm beam profile, performed after a spherical mirror and a
 195 dispersive grating to filter out the 343 nm beam, is reported in
 196 the inset in Fig. 2.

197 We start by investigating PM effects at a driving pulse dura-
 198 tion of 500 fs. Figure 4 shows the experimentally measured
 199 and simulated power of the 114 nm beam as a function of gas
 pressure, for different focused beam sizes and energies. As ob-

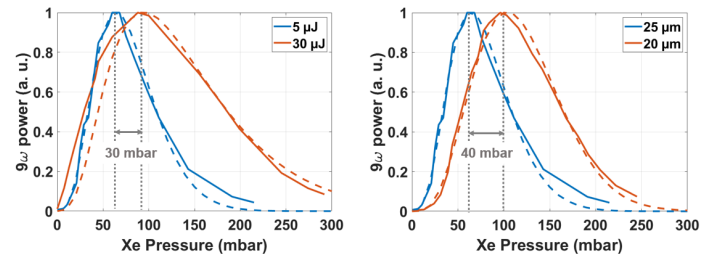


Fig. 4. Measured (solid line) and simulated (dashed line) power at 114 nm as a function of Xe pressure, normalized to the maximum value. Left: the beam diameter is 25 μm (obtained using a lens with a focal length of 200 mm), the pulse energies are 5 μJ (blue) and 30 μJ (red). Right: the input energy is 5 μJ , the focused beam diameters are 20 μm (red) and 25 μm (blue), achieved using lenses with focal lengths of 150 mm and 200 mm, respectively.

200 served in Fig. 4 (right), the PM pressure increases for smaller
 201 focused beam diameter, as expected for example from Eq. (5).
 202 Fig. 4 (left) shows that the PM gas pressure also increases for
 203 increasing input energy, as predicted by Eq. (4), because of the
 204 onset of the ionization, causing a non-negligible contribution of
 205 free electrons. Numerical simulations are in quantitative agree-
 206 ment, reproducing both the PM pressure shifts and changes in
 207 the curve shapes. In Fig. 5, the measured power at 114 nm as
 208 a function of Xe pressure for pulse durations of 100 fs and 3
 209

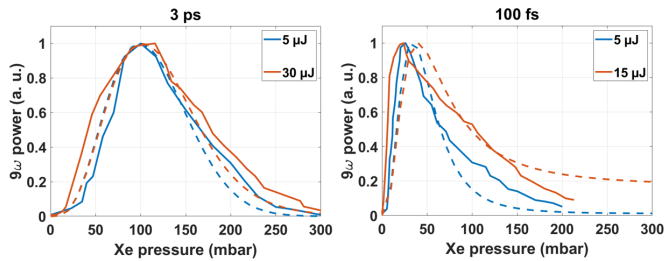


Fig. 5. Measured (solid line) and simulated (dashed line) power at 114 nm as a function of Xe pressure. Left: the pulse duration is 3 ps, the pulse energies are 5 μJ and 30 μJ . Right: the pulse duration is 100 fs, the pulse energies are 5 μJ and 15 μJ .

ps are presented. The focused spot diameter is 38 μm for the 100 fs pulses and 22 μm for the 3 ps pulses, which explains the difference in PM pressure at low pulse energies. In contrast to the data obtained for 500 fs pulses, we do not observe a PM pressure shift as a function of input pulse energy. In both cases, this can be explained by the fact that the available energies and focusing conditions lead to intensities that are below the onset of significant ionization. As discussed above, this intensity threshold is much higher for 100 fs pulses than for 3 ps pulses. The simulations are in reasonable agreement with the observed data.

We now comment on the absolute photon fluxes at 114 nm measured for the three pulse durations. To estimate them, the detected current is converted to photon flux assuming a photoelectric yield for Au of 0.08 electron per photon [22]. Figure 6 shows the photon flux as a function of input pulse energy when the pressure is fixed to the PM pressure for the maximum energy available for each beamline. At a given pulse energy, the intensity is higher for the 100 fs beamline, which translates into a higher efficiency. It is also apparent that the conversion efficiency increases with power for the 100 fs and 3 ps cases, for which ionization effects are small, which is expected for an unsaturated phase-matched third order process. However, for the 500 fs case, this efficiency appears to be almost constant. This could be caused by the fact that ionization effects are local in space and time, resulting in a constant average efficiency as the power is increased. The maximum measured photon fluxes after the LiF window at 100 fs, 500 fs, and 3 ps are 9×10^{12} , 7×10^{12} , and 2×10^{12} ph/s respectively. Taking into account the transmission efficiency of the LiF window which is measured to be 30%, these photon fluxes correspond to optical-to-optical power internal conversion efficiencies of 3×10^{-5} , 1×10^{-5} , and 3×10^{-6} .

In conclusion, we report a VUV table-top source at 114 nm based on cascaded THG of FTL driving pulses at 1030 nm with switchable pulse durations of 100 fs, 500 fs, and 3 ps. Assuming that the spectral widths are maintained during the wavelength conversion stages, as indicated by numerical simulations, this corresponds to energy resolutions of 15, 3, and 0.5 meV. This versatility in terms of temporal and spectral resolution, combined with a high repetition rate and sufficient photon flux should make this type of source very appealing for future tr-ARPES setups in particular in combination with high-luminosity spectrometers [23].

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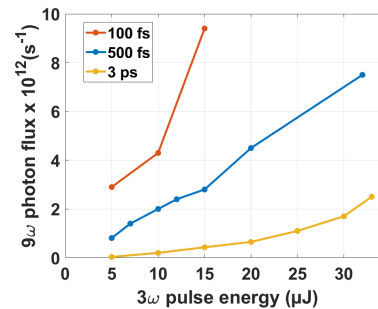


Fig. 6. Estimated photon flux at 114 nm as a function of input power at 343 nm for each pulse duration.

with Z. Zhao, F. Cilento and O. Tjernberg.

Disclosures. The authors declare no conflicts of interest.

Data Availability Statement. Data underlying the results presented in this article are not publicly available at this time but may be obtained from the authors upon reasonable request.

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