Production of narrowband tunable extreme-ultraviolet radiation by noncollinear resonance-enhanced four-wave mixing
Hannemann, S.; Hollenstein, U.; van Duijn, E.J.; Ubachs, W.M.G.

published in
Optics Letters
2005

DOI (link to publisher)
10.1364/OL.30.001494

document version
Publisher's PDF, also known as Version of record

Link to publication in VU Research Portal

citation for published version (APA)

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal

Take down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

E-mail address:
vuresearchportal.ub@vu.nl
Production of narrowband tunable extreme-ultraviolet radiation by noncollinear resonance-enhanced four-wave mixing

S. Hannemann, U. Hollenstein, E.-J. van Duijn, and W. Ubachs

Laser Centre, Department of Physics and Astronomy, Vrije Universiteit, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands

Received December 16, 2004

Fourier-transform-limited extreme-ultraviolet (XUV) radiation (bandwidth ≤300 MHz) tunable around 91 nm is produced by use of two-photon resonance-enhanced four-wave mixing on the Kr resonance at 94 093 cm⁻¹. Noncollinear phase matching ensures the generation of an XUV sum frequency 2ω₁ + ω₂ that can be filtered from auxiliary laser beams and harmonics by an adjustable slit. Application of the generated XUV light is demonstrated in spectroscopic investigations of highly excited states in H₂ and N₂. © 2005 Optical Society of America

OCIS codes: 190.4380, 140.7240, 300.6400.

Nonlinear optical schemes for the production of narrowband and tunable extreme-ultraviolet (XUV) radiation, either through third-order harmonic conversion or resonant sum- and difference-frequency mixing, date back to the 1970s and 1980s. A description of phase matching and properties of the χ(3) nonlinear susceptibility tensor was given by Bjorklund.1 Although Hilber et al.² performed pioneering studies of the resonance enhancement effect by two-photon transitions in noble gases, it was later found that the 4p⁶−5p[1/2]₀ transition in Kr gas is most effective in enhancing the XUV yield.³,⁴ The replacement of grating-based pulsed lasers by Fourier-transform-limited lasers, generally in the form of pulsed-dye amplifiers (PDAs) opened the possibility of generating ultranarrowband XUV radiation⁵,⁶ even at wavelengths as short as 58 nm.⁷ In applications in which the generated XUV beam needs to be separated from the incident laser beams, as well as from the auxiliary harmonic and mixed frequencies, usually a grating is employed, with the drawback of intensity loss of an order of magnitude or more. Here we demonstrate that phase matching in a noncollinear beam configuration can combine the advantage of resonance enhancement with the production of a separated XUV beam, filtered geometrically by the insertion of an adjustable slit. The scheme is somewhat similar to that of BOXCARS, which is applied for geometric filtering of a generated beam in coherent anti-Stokes Raman spectroscopy.⁸

All the measurements are performed in a three-chamber differentially pumped vacuum setup that has been described in connection with third-harmonic generation⁹ (THG) and fifth-harmonic generation.⁷ Here we produce tunable XUV light by mixing the output of two different lasers in a pulsed jet of Kr. One provides the resonant light ω₁ for the Kr 4p⁶−5p[1/2]₀ two-photon transition at 94 093 cm⁻¹; the other supplies the tunable component ω₂ in a resonance-enhanced four-wave-mixing scheme. For all the results presented, the sum frequency 2ω₁ + ω₂ is used, and the repetition rate is 10 Hz. To filter out undesired wavelengths, a simple geometric scheme is applied as depicted in Fig. 1. Aligning the resonant light ω₁ at 212 nm and the tunable output ω₂ of the second laser to overlap in the Kr jet under an angle of approximately 80 mrad allows phase-matching conditions for the sum frequency 2ω₁ + ω₂, the difference frequency 2ω₁ − ω₂, and the third harmonic 3ω₁ to be fulfilled at angles as displayed in Fig. 1. This geometry allows us to block all unwanted wavelengths with a slit that is adjustable in width and position and located between the frequency-mixing chamber and the downstream application zone.

For the production of the resonant light at 212 nm a novel narrowband laser system, as schematically shown in Fig. 2, is used. A gain-switched injection-seeded Ti:sapphire (Ti:Sa) oscillator, pumped by the second-harmonic output of a Q-switched Nd:YAG laser (Spectra-Physics Quanta Ray GCR-3), produces nearly Fourier-transform-limited pulses at a wavelength of λ=850.222 nm with a typical duration of 15 ns and a bandwidth of 40 MHz. The output of the pulsed oscillator is subsequently enhanced to 15 mJ

Fig. 1. Schematic view of the beam alignment and the resulting directions for the different vacuum ultraviolet wavelengths. In the Kr gas jet the sum frequency 2ω₁ + ω₂, difference frequency 2ω₁ − ω₂, and third-harmonic 3ω₁ are produced. A phase-matching diagram is included in the upper left. The slit, adjustable in position and width, filters the XUV beam at frequency 2ω₁ + ω₂.
Difference with results of Namioka.  

Drive the two-photon transition in Kr, the error multiplied by the fourth-harmonic wavelength-meter, which is 0.002 cm\(^{-1}\) in the near-infrared. Since the fourth-harmonic wavelength-meter is locked to a preset value of an ATOS wavelength meter. For all the H\(_2\) lines observed vibration procedure for terms of Abgrall et al.  

in a bow-tie Ti:Sa amplifier and nonlinearly upconverted by two consecutive type I second-harmonic generation stages made from \(\beta\)-barium borate (BBO) crystals cut at appropriate phase-matching angles. The final output at 212 nm is typically 0.5 mJ with a bandwidth of 80 MHz. The injection-seeding light is provided by an external grating diode laser system (Toptica DL 100). The output frequency of the diode laser is locked with a computer-controlled feedback loop. The computer continuously acquires the wavelength of the seed light from an ATOS wavelength-meter and controls the output wavelength of the diode laser by adjusting the angle of the feedback grating with a piezoactuator. The accuracy of this locking scheme is restricted by the accuracy of the wavelength-meter, which is 0.002 cm\(^{-1}\) in the near-infrared. Since the fourth-harmonic \(\omega_2\) is used to drive the two-photon transition in Kr, the error multiplies by 8 to 0.016 cm\(^{-1}\) in the XUV. It should be noted that under conditions of f=25 cm focusing of the \(\omega_1\) beam the two-photon resonance in Kr broadens to approximately 1 cm\(^{-1}\). In view of the small bandwidth of the \(\omega_1\) laser (=0.003 cm\(^{-1}\) at 212 nm), the resulting XUV frequency and bandwidth are determined by the frequency and bandwidth of the laser, not by the resonance.

Tunable frequency calibration of the PDA (bandwidth of \(\approx\)100 MHz) is realized through iodine saturation spectroscopy and a stabilized etalon yielding an accuracy in \(\omega_2\) (including frequency chirp effects in the PDA) better than 0.001 cm\(^{-1}\). The intensity of the XUV output was not measured directly. From measurements of signal strengths in photoionization studies (see below) and comparison with previous detection schemes using THG,9–11 photon densities in excess of 10\(^8\) photons per pulse are estimated.

To demonstrate the capabilities of the source, the tunable narrowband XUV radiation is applied in crossed-molecular-laser-beam spectroscopic experiments on highly excited states in H\(_2\) and N\(_2\). Spectral lines are recorded by tuning the XUV source into resonance with an excited state of the molecule, which is then subsequently ionized (a 1+1’ photoionization scheme) by an auxiliary laser beam—here the residual second harmonic of the Ti:Sa laser at 425 nm is taken—incident from the rear side of the interaction chamber. The signal is monitored by detection of ions on an electron multiplier after pulsed-field extraction, time-of-flight mass separation, and time gating of the ions produced in photoionization. We reduce Doppler effects (broadening and shifts) by producing a collimated molecular beam by means of a skimmer and aligning it perpendicularly to the resulting (2\(\omega_1\) + \(\omega_2\)) XUV beam. We accomplish the latter by adjusting the angles between incident laser beams and by addressing shifts between spectral lines in pure H\(_2\) beams and seeded H\(_2\)/Ar beams.

For H\(_2\), measurements are performed on the B\(^1\Sigma^+_u\) – X\(^1\Sigma^+_g\) (19,0) Lyman band; results for the line positions after averaging over various recordings and assessment of the uncertainty budget (dominated by the uncertainty in \(\omega_1\)) are listed in Table 1. An error of 0.020 cm\(^{-1}\) is estimated, except for the P(3) line for which no saturated I\(_2\) line was available in the calibration procedure for \(\omega_2\); here the calibration relied on a wavelength meter. For all the H\(_2\) lines observed

Table 1. Observed Transition Frequencies for P and R Branch Lines in the H\(_2\)B\(^1\Sigma^+_u\) – X\(^1\Sigma^+_g\) (19, 0) Lyman Band\(^a,b\)

<table>
<thead>
<tr>
<th>(J)</th>
<th>(P(J))</th>
<th>(\Delta_1)</th>
<th>(\Delta_2)</th>
<th>(\Delta_3)</th>
<th>(\Delta_4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>109 653.067(20)</td>
<td>-0.17</td>
<td>0.10</td>
<td>-0.01</td>
<td>-0.05</td>
</tr>
<tr>
<td>2</td>
<td>109 436.712(20)</td>
<td>-0.23</td>
<td>-0.34</td>
<td>-0.17</td>
<td>-0.07</td>
</tr>
<tr>
<td>3</td>
<td>109 124.687(40)</td>
<td>-0.16</td>
<td>-0.25</td>
<td>0.22</td>
<td>-0.12</td>
</tr>
</tbody>
</table>

\(^a\Delta_1\) represent deviations from previous investigations (present minus previous).

\(^b\)All values given in cm\(^{-1}\).

\(^c\)Difference with the results of Hinnen et al.10

\(^d\)Difference with results of Namioka.12

\(^e\)Difference with results of Abgrall et al.13

\(^f\)Difference with the calculated line positions resulting from the term levels of Abgrall et al.14 and the ground-state level energies from Jennings et al.15


Fig. 2. Laser setup for the resonant light at 212 nm. In a computer-controlled feedback loop the diode injection seed is locked to a preset value of an ATOS wavelength meter.

Fig. 3. 1+1’ photoionization recording of the R (0–2) lines of the b’’\(\Sigma^+\) \(\rightarrow\) X\(^1\Sigma^+_g\) (8, 0) band in N\(_2\).
Deviations from results of previous investigations due to larger bandwidths,\textsuperscript{16} from old classical spectroscopic studies with narrowband pulsed molecular beam. In Table 1 the observed broadening in the slightly divergent high-velocity line in the $\Sigma^+_u$, $X^1\Sigma^+_g^+$ bands of H\textsubscript{2} and N\textsubscript{2}.\textsuperscript{17,18} The observed linewidth of $\approx$600–700 MHz was observed. This is the same width as obtained in studies investigating H\textsubscript{2} spectral lines with narrowband XUV produced through direct THG. From a comparison with results from Ref. 11 we estimate that the bandwidth of the $2\omega_2+\omega_1$ XUV beam is $\approx$300 MHz. The additional broadening is due to residual Doppler broadening in the slightly divergent high-velocity pulsed molecular beam. In Table 1 the observed transition frequencies are compared with previous results from XUV laser spectroscopy, however, at larger bandwidths,\textsuperscript{10} from old classical spectroscopic studies\textsuperscript{12} and from a recent highly accurate classical study from the Meudon group.\textsuperscript{13,14} Very accurate data result from Meudon excited-state level energies\textsuperscript{14} combined with ground-state level energies from far-infrared spectra;\textsuperscript{15} these data are off by only $\pm$0.06 cm$^{-1}$ from the present data, which represent the highest accuracy frequencies on the (19,0) Lyman band. These lines could be included in analyses of quasar data aiming at uncovering a possible variation of the proton-to-electron mass ratio over cosmological time.\textsuperscript{11}

XUV spectroscopic investigations in N\textsubscript{2} were performed as well. A single rotationally resolved $Q(1)$ line in the $b^1\Pi_u - X^1\Sigma_g^+$ (12, 0) band was recorded, for which a linewidth of $\Gamma_{\text{obs}}=1.74\,(15)$ GHz was observed. After deconvolution of contributions from Doppler and laser source broadening a value results for the natural width, associated with the predissociation rate of the excited state; an excited lifetime of $t_{\text{exc}}=105\,(25)$ ps is estimated. In previous laser-based studies with larger bandwidths\textsuperscript{16} a natural lifetime broadening effect could not be discerned. In addition, the $b^1\Sigma_u^+ - X^1\Sigma_g^+$ (8, 0) band was recorded, for the first time to our knowledge resolving the band head section, as shown in Fig. 3. The obtained width of 750 MHz is somewhat broader than the expected instrument width and hence indicates predissociation of the $b^1\Sigma_u^+-v=8$ upperstate, but at a lower rate than for $b^1\Pi_u,-v=12$. In Table 2 results on calibrated frequencies and observed widths are listed. For the measured transition frequencies a comparison is made with values obtained from previous studies.\textsuperscript{17,18}

In conclusion, narrowband XUV generation using a noncollinear phase-matching scheme for resonance-enhanced four-wave mixing is demonstrated and applied in spectroscopic measurements on highly excited states of H\textsubscript{2} and N\textsubscript{2}. The accuracy of the obtained results is dominantly limited by the calibration of the $\omega_1$ frequency laser and could be improved considerably when a better calibration procedure becomes available.

The authors thank J. P. Sprenger and E. Reinhold (Laser Centre Vrije Universiteit), as well as F. Merkt (ETH Zürich), for helpful discussions. We gratefully acknowledge financial support from the Netherlands Foundation for Fundamental Research of Matter. S. Hannemann’s e-mail address is sandro@nat.vu.nl.

### References


### Table 2. Observed Transition Frequencies and Linewidths $\Gamma_{\text{obs}}$ for Lines in the $b^1\Pi_u - X^1\Sigma_g^+$ (12, 0) Band and the $b^1\Sigma_u^+ - X^1\Sigma_g^+$ (8, 0) Band in N\textsubscript{2}$^a$

<table>
<thead>
<tr>
<th>Line</th>
<th>Position</th>
<th>$\Delta$</th>
<th>$\Gamma_{\text{obs}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$b^1\Pi_u - X^1\Sigma_g^+$ (12, 0)</td>
<td>$Q(1)$</td>
<td>109 829.483(30)</td>
<td>$-0.10^c$</td>
</tr>
<tr>
<td>$b^1\Pi_u - X^1\Sigma_g^+$ (8,0)</td>
<td>$R(1)$</td>
<td>109 546.800(20)</td>
<td>$-0.23^d$</td>
</tr>
<tr>
<td>$b^1\Pi_u - X^1\Sigma_g^+$ (8,0)</td>
<td>$R(0)$</td>
<td>109 546.211(20)</td>
<td>$-0.19^d$</td>
</tr>
</tbody>
</table>

\textsuperscript{a}All values given in cm$^{-1}$

\textsuperscript{b}Deviations from results of previous investigations (present minus previous).

\textsuperscript{c}Roncin et al.\textsuperscript{18}

\textsuperscript{d}Carroll et al.\textsuperscript{17}

\textsuperscript{e}Ubachs et al.\textsuperscript{16}