Photon-matter interaction at short wavelengths and ultra-high intensity – Gas-phase experiments at FLASH

2008 J. Phys.: Conf. Ser. 141 012014

(http://iopscience.iop.org/1742-6596/141/1/012014)

Journal of Physics: Conference Series **141** (2008) 012014 doi:10.1088/1742-6596/141/1/012014

Photon-matter interaction at short wavelengths and ultra-high intensity – Gas-phase experiments at FLASH

M Richter[1], S V Bobashev[2], A A Sorokin[1,2] and K Tiedtke[3]

[1] Physikalisch-Technische Bundesanstalt, Abbestraße 2-12, 10587 Berlin, Germany

[2] Ioffe Physico-Technical Inst., Polytekhnicheskaya 26, 194021 St. Petersburg, Russia

[3] Deutsches Elektronen-Synchrotron, Notkestraße 85, 22603 Hamburg, Germany

E-mail: mathias.richter@ptb.de

Abstract. In different focused beams at the new soft X-ray Free-electron LASer in Hamburg FLASH, ion time-of-flight spectroscopy on gas targets was performed. Irradiation levels above 10^{13} up to 10^{16} W cm^{-2} were achieved in the vacuum and extreme ultra-violet. The first group of experiments was performed on nitrogen molecules and neon and helium atoms at wavelengths around 30 nm, i.e. at photon energies around 40 eV. Absolute cross sections for one- and two-photon ionization could be derived. The second group of experiments was performed on different rare gases at the wavelength of 13.3 nm, i.e. at the photon energy of 93 eV. As an example, the generation of Xe^{21+} was observed which requires a total energy of at least 5 keV absorbed per atom, starting from neutral Xe, within the FLASH pulse duration of about 10 fs. Here, the situation might be beyond the multiphoton scheme and perturbation theory.

1. Introduction

The upcoming laser facilities like the Free-electron LASer in Hamburg FLASH, the Linac Coherent Light Source (LCLS) in Stanford, the Spring-8 XFEL in Japan, or the European XFEL in Hamburg are characterized by photon pulses that are sufficiently intense for pulse-resolved studies of materials. In particular, the dynamics of chemical reactions on surfaces or within biological systems are scheduled to be investigated by microscopic snapshots using the femtosecond pulses of a focused Free-Electron Laser (FEL) beam at short wavelengths [1]. FLASH already provides pulse energies up to 100 µJ at pulse durations of 10 to 50 fs and, hence, peak power values of more than 1 GW in the wavelength range from 50 down to 6.5 nm [2,3]. In focused beams, irradiation levels above 10^{13} up to 10^{16} W cm^{-2} have recently been achieved [4-9]. Here, photon-matter interaction is affected by nonlinear processes whose mechanisms seem to differ in some respects from those which are known from optical radiation. Thus, studies on photon-matter interaction at short wavelengths and ultra-high intensity are of general significance for the interpretation of any experimental result of materials research obtained with the current and future X-ray lasers. In order to investigate nonlinear processes within an atom or molecule, such as multiphoton excitation and ionization, gas-phase experiments are well suited because the influence from next neighbours may be detected by varying the gas pressure and can largely be suppressed by working at sufficiently low target density. At FLASH, first experiments on molecular nitrogen and rare gases were performed by ion Time-Of-Flight (TOF) mass/charge spectroscopy [4-6], electron spectroscopy [7], and ion recoil momentum spectroscopy using a so-

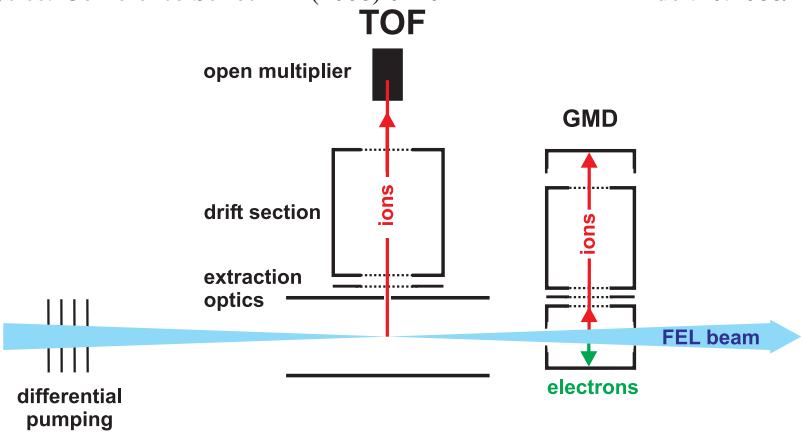

Figure 1. Schematic diagram of the experimental setup as used for ion Time-Of-Flight (TOF) spectroscopy on gas targets at the microfocus beamline BL2 of the soft X-ray Free-Electron Laser (FEL) FLASH using a Gas-Monitor Detector (GMD) for the online detection of absolute pulse energies [9,10].

called reaction microscope [8]. In the present paper, multiphoton and strong-field phenomena are discussed which were quantitatively studied at FLASH on free atoms and molecules by means of ion TOF spectroscopy at wavelengths around 30 and at 13.3 nm.

2. Multiphoton ionization in the vacuum ultra-violet

Figure 1 shows a scheme of the experimental setup as used in the 15-µm focus (FWHM) of the beamline BL2 at FLASH in the wavelength range from 29 to 33 nm, i.e. in the photon energy range from 38 to 43 eV [9]. FEL pulses with a pulse duration of $\Delta t = (25 \pm 5)$ fs [2] were introduced into a gas target at pressures from 10^{-4} to 10^{-3} Pa. Generated photoions were extracted towards the TOF spectrometer entrance aperture by an homogenous electric field. The aperture width of 1 mm along the photon beam ensured photoion collection exclusively from the focus region with a Rayleigh lengths larger than 1 mm. The ions were detected by means of an open multiplier whose efficiency was tested to be linear in the range from 1 to 10^6 particles detected per FEL pulse. The whole apparatus could be moved ± 2 cm along the photon beam in order to vary the beam cross section A and, thus, the mean irradiance $E = W\,\Delta t^{-1}A^{-1}$ within the interaction volume accepted by the TOF spectrometer. Online detection of absolute pulse energy W and the photon number per FEL pulse N_{ph} was performed about 50 cm behind the focal region by using a calibrated Gas-Monitor Detector (GMD) [10] profiting from the high transparency of the target gas at the working pressures.

Figure 2(a) shows a single-shot TOF spectrum of molecular nitrogen (N_2) taken at the photon energy of $\hbar\omega = 38.0$ eV from a series of FEL pulses with a distance of 1 µs [4]. Besides the strong N_2^+ peaks, signals due to the generation of N_2^{++} ions and/or N^+ dissociation fragments, which have the same mass/charge ratio, arise at about 1.5 µs lower time-of-flight, respectively. Their multiplet structure, more evident in the inset, is explained by Coulomb repulsion of N^+ fragments. The local maxima at the edges of the multiplet, with less emission in the centre, indicate dominance of those dissociation channels with two repulsive N^+ fragments generated simultaneously which influences their time-of-flight, respectively. In a recent study performed at the SPring-8 XUV-FEL with 24.6 eV photons, these side peaks are less pronounced compared to the emission in the centre [11].

However, even our photon energy of 38.0 eV was not sufficient to simultaneously generate two N^+ fragments from N_2 so that a multiphoton process must be suspected [4,11]. This assumption is confirmed by the dependence of the measured (N_2^{++}& N^+) to N_2^+ ion-yield ratio on the photon exposure N_{ph}/A or irradiance $E = (N_{ph}/A) \times (\hbar\omega/\Delta t)$ as shown in figure 2(b). The almost constant value of 0.23 for this ratio at photon exposures below 2×10^{16} cm^{-2} is explained by the linear one-photon processes $N_2 \rightarrow (N^+ + N)$ and $N_2 \rightarrow N_2^+$ [12]. At $N_{ph}/A = 2 \times 10^{16}$ cm^{-2}, the mean number of N_2^+ ions generated within our interaction volume per FEL pulse $N(N_2^+)$ can be estimated, using the $N_2 \rightarrow N_2^+$ photoionization cross section $\sigma_1 = 10.8 \times 10^{-18}$ cm^2 [12], according to:

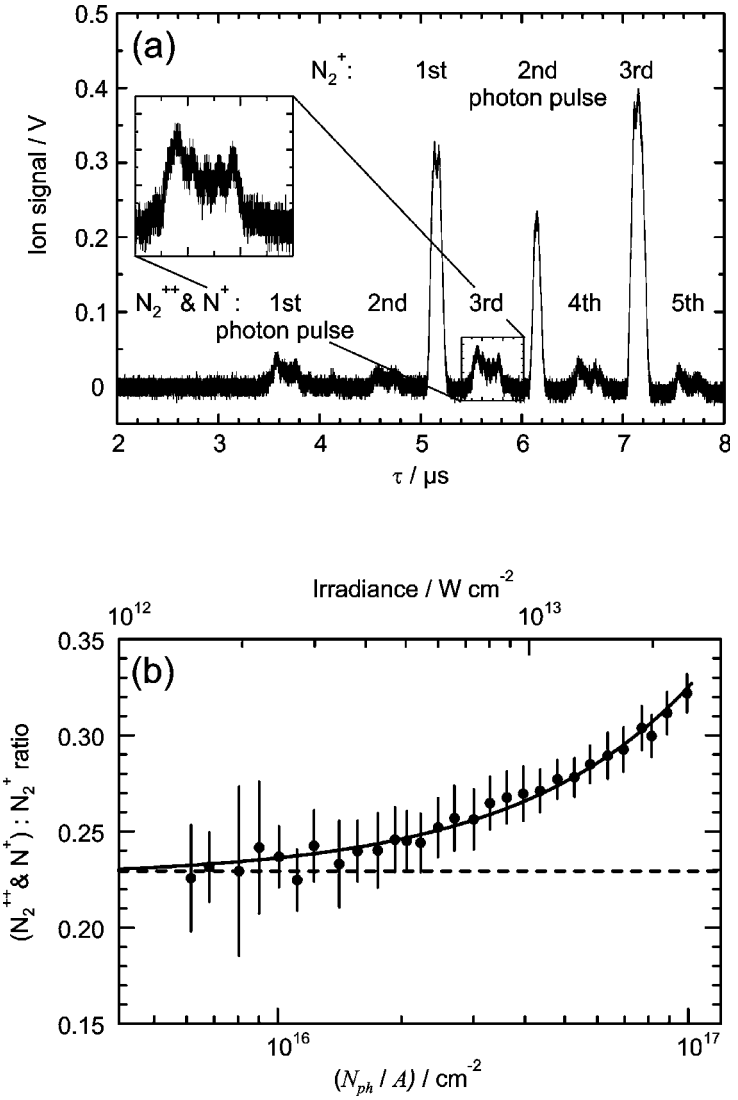

Figure 2. (a) Single-shot ion TOF spectrum of molecular nitrogen (N_2) obtained from a series of subsequent FEL pulses separated by 1 μs. The inset shows an enlargement of the N_2^{++} and N^+ signals from the third pulse. (b) Measured ratio for the number of N_2^{++} and N^+ ions versus the number of N_2^+ ions generated per photon pulse as a function of absolute photon exposure N_{ph}/A at the photon energy of $\hbar\omega$ = 38.0 eV. The solid line represents a linear fit curve according to equation (3). The upper irradiance scale was obtained according to $E = (N_{ph}/A) \times (\hbar\omega/\Delta t)$ assuming a photon-pulse duration of Δt = 25 fs [4].

$$\frac{N(N_2^+)}{N(N_2)} \approx \sigma_I \frac{N_{ph}}{A} \tag{1}$$

and amounts already to about 20 % of the number of initial N_2 targets $N(N_2)$. Above 2×10^{16} cm^{-2}, thus, further ionization and fragmentation of the ions as intermediate targets is, in fact, expected within the scheme of a sequential two-photon double ionization:

$$N_2 \xrightarrow{\text{1st photon}} N_2^+ \xrightarrow{\text{2nd photon}} N^+ + N^+ \qquad (2)$$

to nonlinearly contribute to the (N_2^{++}& N^+) generation. As a result of solving the rate equations for this two-photon process, the (N_2^{++}&N^+) to N_2^+ ion-yield ratio should increase with N_{ph}/A according to [4]:

$$\frac{N\left(N_2^{++}\ \&\ N^+\right)}{N\left(N_2^+\right)} \approx 0.23 + \sigma_{II}\frac{N_{ph}}{A} \qquad (3)$$

with the cross section σ_{II} for the second step, i.e. for the single photoionization and fragmentation of the intermediate N_2^+ targets. Fitting our data in figure 2(b) by the expression in equation 3 and neglecting a non-fragmenting $N_2^+ \rightarrow N_2^{++}$ process for the second step in equation 2, yields a value for σ_{II} in the order of $(1.0 \pm 0.3) \times 10^{-18}$ cm^2. For this purpose, N_{ph} was measured by means of the GMD and A was determined via signal saturation due to target depletion [9].

Our experimental results on nitrogen molecules demonstrate that (soft) X-ray lasers like FLASH allow the investigation of ions that are prepared as intermediate targets from initial atomic or molecular targets in a first step of a photoionization sequence according to equation 2 by the highly intense photon pulses themselves. By corresponding experiments on neon (Ne) atoms, this idea was applied to measure also the one-photon single ionization cross section of $Ne^+ \rightarrow Ne^{2+}$ (second step in figure 3(b)) and the direct two-photon single ionization of $Ne^{2+} \rightarrow Ne^{3+}$ (third step in figure 3(c)) at 42.8 eV photon energy, respectively, and the direct or resonant two-photon single ionization cross section of $Ne^+ \rightarrow Ne^{2+}$ at 38.4 eV photon energy (second step in figure 3(d)) [5]. The data were derived from the dependence of the Ne^{2+} to Ne^+ and Ne^{3+} to Ne^{2+} ion-yield ratios on photon exposure and irradiance as shown in figure 4. For this purpose, the data points were fitted by applying the general power law of perturbation theory for the rate of n-photon processes:

$$\dot{N}^{(n)} = N\,\sigma^{(n)}\left(\frac{E}{\hbar\omega}\right)^n \qquad (4)$$

with the respective generalized n-photon ionization cross sections $\sigma^{(n)}$ and the number N of (intermediate) targets within the interaction volume.

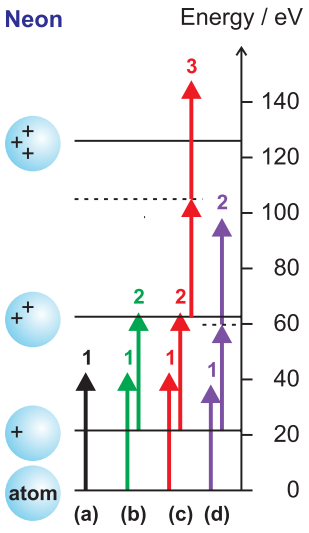

Figure 3. Energy diagram [13] and photoionization schemes for neon: one-photon single ionization (a), sequential two-photon double ionization (b), and four-photon triple ionization (c) by 42.8 eV photons and three-photon double ionization (d) by 38.4 eV photons. The dashed lines indicate virtual and/or resonant states.

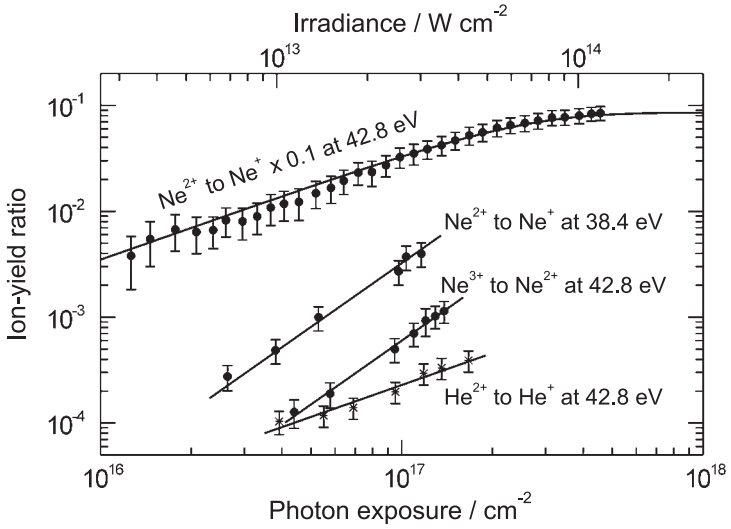

Figure 4. Ion-yield ratios as a function of mean pulse irradiance and photon exposure measured at 42.8 and 38.4 eV photon energy [5].

All cross section values we have derived from our gas-phase photoionization experiments at FLASH around 40 eV photon energy are listed in table 1. It comprises also the value of $(1.6 \pm 0.6) \times 10^{-52}$ cm^{4}s for the direct two-photon double ionization cross section of He → He^{2+} at 42.8 eV photon energy, evaluated from the measured He^{2+} to He^+ ion-yield ratio which is shown in figure 4, too. The latter process has been of considerable interest during the last years demonstrated by numerous theoretical predictions of the corresponding cross section (see, e.g., [14,15] and references therein). Best agreement with our experimental result is achieved, so far, by a recent calculation by Ivanov and Kheifetz which yields $(1.3 \pm 0.3) \times 10^{-52}$ cm^{4}s at 42.5 eV photon energy [14].

Table 1. Cross section values measured for different one- and two-photon ionization processes on atomic, molecular, and ionic targets [4,5].

Photoionization process	Cross section
One-photon single ionization/dissoziation of N_2^+ at 38.0 eV photon energy	$(1.0 \pm 0.3) \times 10^{-18}$ cm^2
One-photon single ionization of Ne^+ at 42.8 eV photon energy	$(7.0 \pm 1.0) \times 10^{-18}$ cm^2
Two-photon single ionization of Ne^+ at 38.4 eV photon energy	$(1.6 \pm 0.6) \times 10^{-50}$ cm^{4}s
Two-photon single ionization of Ne^{2+} at 42.8 eV photon energy	$(3.0 \pm 1.2) \times 10^{-51}$ cm^{4}s
Two-photon double ionization of He at 42.8 eV photon energy	$(1.6 \pm 0.6) \times 10^{-52}$ cm^{4}s

3. Multiphoton ionization and strong-field phenomena in the extreme ultra-violet

In comparison to the FLASH experiments in the Vacuum Ultra-Violet (VUV) described, the main difference in the experimental geometry for our measurements in the Extreme Ultra-Violet (EUV) at the wavelength of 13.3 nm, i.e. at the photon energy of 93 eV, concerns the FEL beam focusing as demonstrated by figure 5. Profiting from optical technology development in the field of EUV Lithography (EUVL), a spherical Mo-Si multilayer mirror with a reflectance in the order of 68% was used under normal incidence and a microfocus of 3 μm in diameter (FWHM) could be generated in back-reflexion geometry [6]. As a result, the irradiance levels achieved were by about two orders of

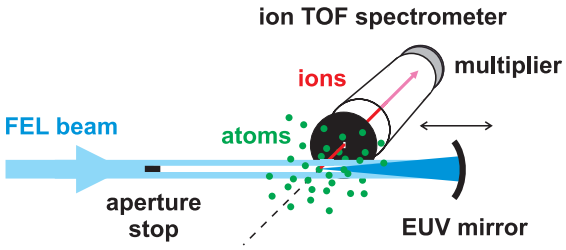

Figure 5. Experimental setup for the investigation of atoms by ion time-of-flight (TOF) spectroscopy in a Free-Electron Laser (FEL) focus of a spherical Extreme Ultra-Violet (EUV) multilayer mirror [6].

magnitude higher than during our VUV measurements. In order to prevent ion signals due to the incident unfocused radiation, an aperture stop of 1.5 mm in height was used in combination with a spectrometer entrance aperture of 1.0 mm in height. In stead of the apparatus, here, the mirror was moved along the photon beam to vary beam cross section and irradiance within the interaction volume.

Figure 6 shows ion TOF spectra of Ne obtained (a) in the focus at an irradiance level of 6×10^{15} W cm^{-2} and, for comparison, (b) out of the focus at 2×10^{12} W cm^{-2}. Obviously, the dominating Ne^+ and the weak Ne^{2+} processes that occur already at low irradiance (figure 6b) are due to one-photon single and double ionization whereas the main part of the Ne^{2+} signal and all signals from higher charges that arise in the focus only (figure 6a) seem to be due to nonlinear multiphoton ionization. From the energy diagram in figure 3, one can see that at 93 eV photon energy, a Ne^{2+} state may be reached starting from the Ne^+ ground state and Ne^{3+} from Ne^{2+} via one-photon processes, respectively, as illustrated by the sequential multiphoton scheme in figure 7. However, the generation of Ne^{4+}, Ne^{5+}, and Ne^{6+} starting from the ground state of the preceding charge, respectively, requires already the energy of two EUV photons. Three EUV photons are needed for a corresponding $Ne^{6+} \rightarrow Ne^{7+}$ process. Within the framework of such a sequential multiphoton scheme, Ne^{7+} is explained, overall, by a twelve-photon process. On the other hand, the total energy which must have been absorbed by an individual atom within a single FLASH pulse of 10 fs duration [3] to reach Ne^{7+} from the atomic ground state of Ne amounts to 715 eV [13], i.e. nearly the energy of eight EUV photons.

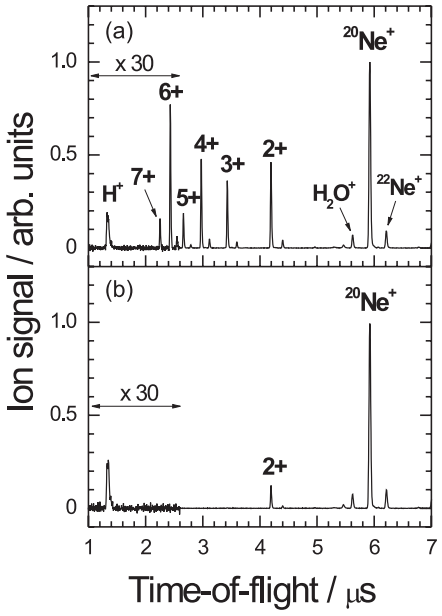

Figure 6. Ion TOF spectra of neon (Ne) taken at 93 eV photon energy and the irradiance levels of 6×10^{15} W cm^{-2} (a) and 2×10^{12} W cm^{-2} (b).

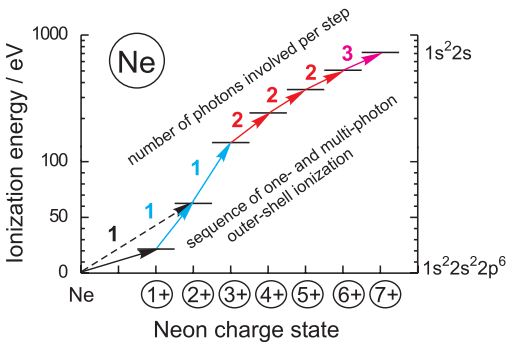

Figure 7. Energy levels and sequential multiphoton ionization channels via ionic states for neon (Ne) at the photon energy of 93 eV.

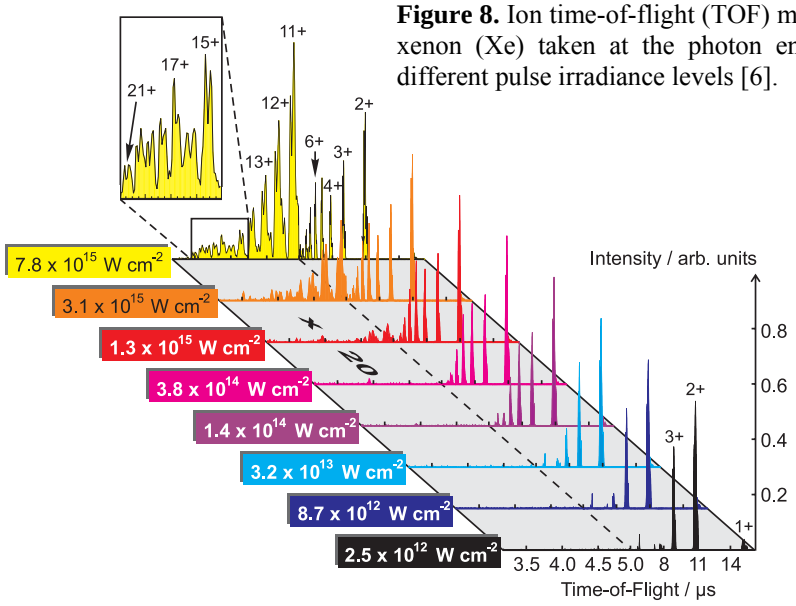

Figure 8. Ion time-of-flight (TOF) mass/charge spectra of xenon (Xe) taken at the photon energy of 93 eV and different pulse irradiance levels [6].

Even more complex is the situation in the case of xenon (Xe) [6]. Figure 8 shows the nonlinear appearance of higher charge states with increasing irradiance E. At $E = 7.8 \times 10^{15}$ W cm^{-2}, Xe^{21+} occurs whose generation, starting from neutral Xe, requires a total energy of at least 5 keV. It means that almost 60 EUV photons at the energy of 93 eV must have been absorbed by an individual atom during a single FEL pulse. A satisfying *ab initio* calculation of these findings within the framework of high-order perturbation theory and a multiphoton scheme is hard to imagine. On the other hand, also semi-classical models within the wave picture of light, as developed for strong-field phenomena in the optical regime [16,17], seem to fail. For their application, the so-called ponderomotive energy which is the quiver energy transferred to a free electron by the oscillating field, is by far too low at so high frequencies.

References

[1] Bostedt C *et al* 2008 *Kai Siegbahn Memorial Volume of Nucl. Instr. and Meth. A*
[2] Ayvazyan V *et al* 2005 *Eur. Phys. J.* D **37** 297
[3] Ackermann W *et al* 2007 *Nat. Photonics* **1** 336
[4] Sorokin A A *et al* 2006 *J. Phys. B: At. Mol. Opt. Phys.* B **39** L299
[5] Sorokin A A *et al* 2007 *Phys. Rev.* A **75** 051402(R)
[6] Sorokin A A *et al* 2007 *Phys. Rev. Lett.* **99** 213002
[7] Nagasono M *et al* 2007 *Phys. Rev.* A **75** 051406(R)
[8] Moshammer R *et al* 2007 *Phys. Rev. Lett.* **98** 203001
[9] Sorokin A A *et al* 2006 *Appl. Phys. Lett.* **89** 221114
[10] Richter M *et al* 2003 *Appl. Phys. Lett.* **83** 2970
[11] Sato T *et al* 2008 *Appl. Phys. Lett.* **92** 15410
[12] Samson J A R, Masuoka T, Pareek P N and Angel G C 1987 *J. Chem. Phys.* **86** 6128
[13] Kelly R L 1987 *J. Phys. Chem. Ref. Data* **16**, Suppl. 1 & 2
[14] Ivanov I A and Kheifets A S 2008 *J. Phys. B: At. Mol. Opt. Phys.* **41** 095002
[15] Feist J *et al* 2008 *Phys. Rev.* A **77** 043420
[16] Protopapas M, Keitel C H and Knight P L 1997 *Rep. Prog. Phys.* **60** 389
[17] Delone N and Krainov V 2000 *Multiphoton Processes in Atoms* (New York: Springer)